Electronic Supplementary Information (ESI)

A general quantitative pH sensor developed with dicyandiamide *N*-doped high quantum yield graphene quantum dots †

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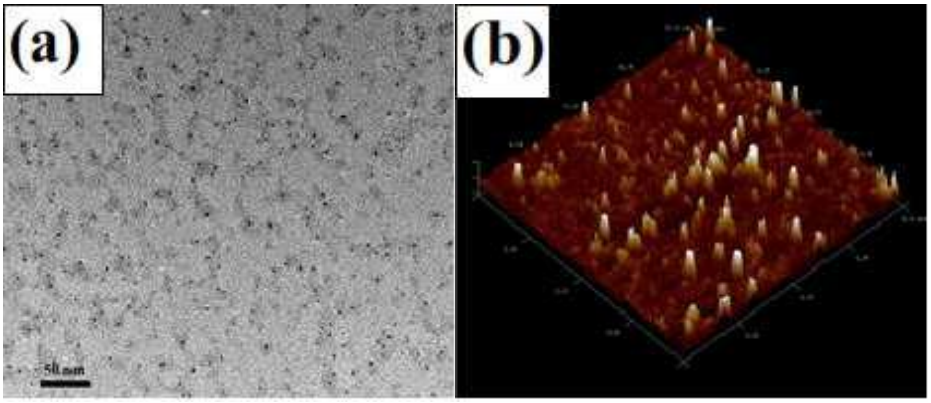


Fig.S1 (a) The TEM and (b) the three-dimension AFM image of as-prepared GQDs.

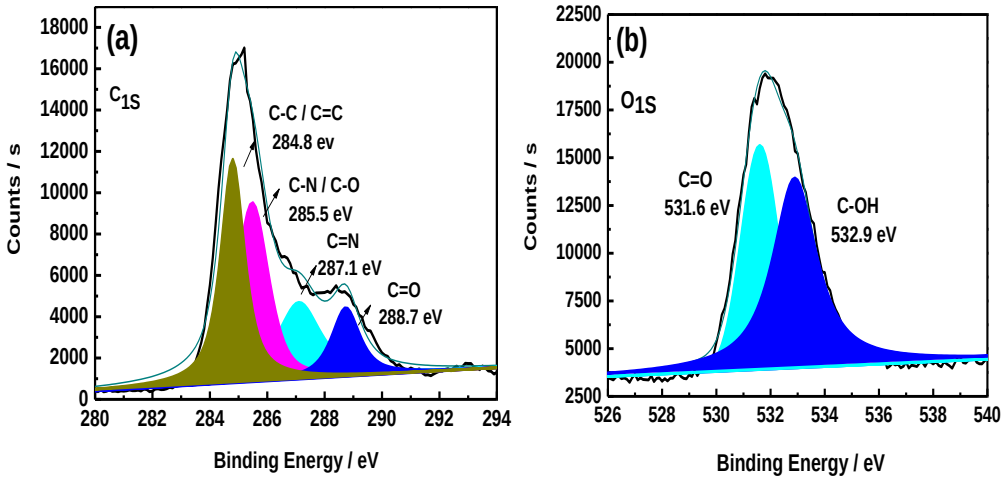


Fig.S2 (a) C 1s, (b) O 1s spectra of GQDs.

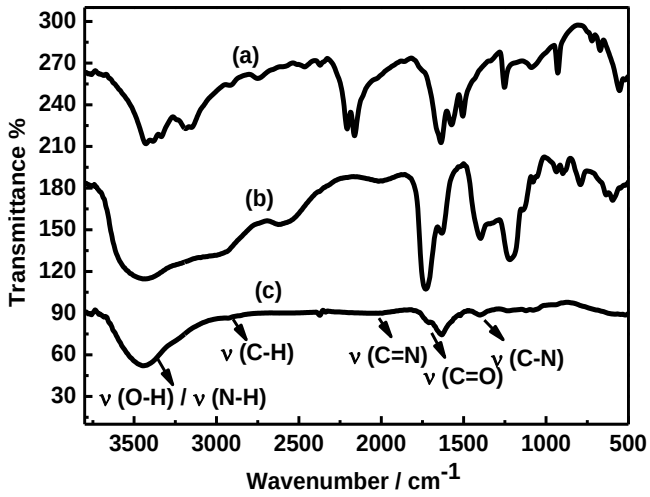


Fig.S3 The Fourier transformed infrared spectrum of (a) dicyandiamide; (b) citric acid; (c) GQDs.

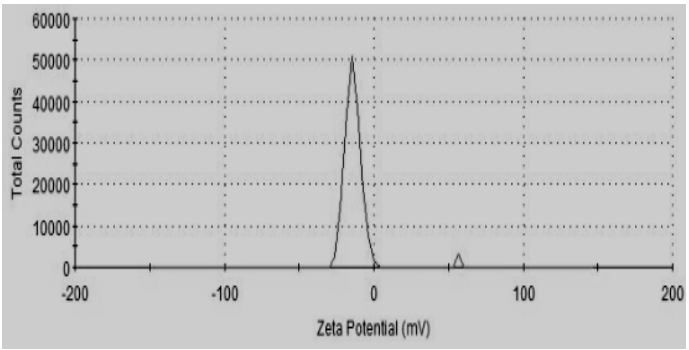


Fig.S4 The zeta potential of the GQDs.

Quantum Yield Calculations

The quantum yield (Φ) of GQDs was calculated using quinine sulfate as reference. Quinine (literature Φ = 0.54) was dissolved in 0.1 M H_2SO_4 (refractive index (η) of 1.33) while the GQDs was dissolved in ultra-pure water (η = 1.33). Then the quantum yield of GQDs was calculated by comparing the integrated photoluminescence intensities (excited at 370 nm) and the absorbance values (at 370 nm) of GQDs with the references Fluorescein sodium. The data was plotted (Figure. S4) and the slopes of the sample and the standards were determined. The data showed good linearity.

The quantum yield was calculated using the below equation:

$$\Phi x = \Phi_{ST} (m_x / m_{ST}) (\eta_x^2 / \eta_{ST}^2)$$

Where Φ is the quantum yield, m is slope, η is the refractive index of the solvent, ST is the standard and X is the sample. The quantum yield for GQDs is 36.5%.

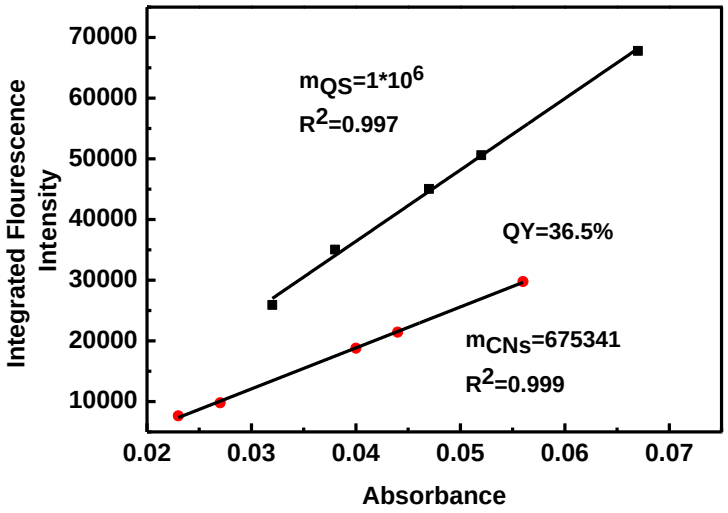


Fig.S5 Plot of integrated photoluminescence intensity vs. absorbance of the GQDs and quinine sulfate.

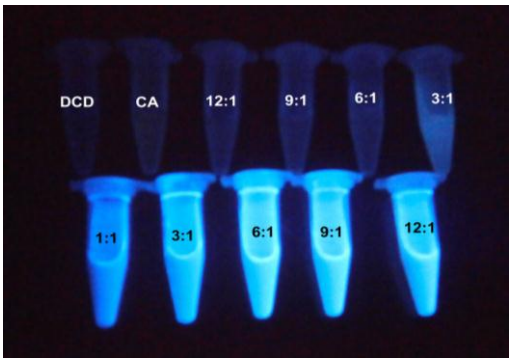
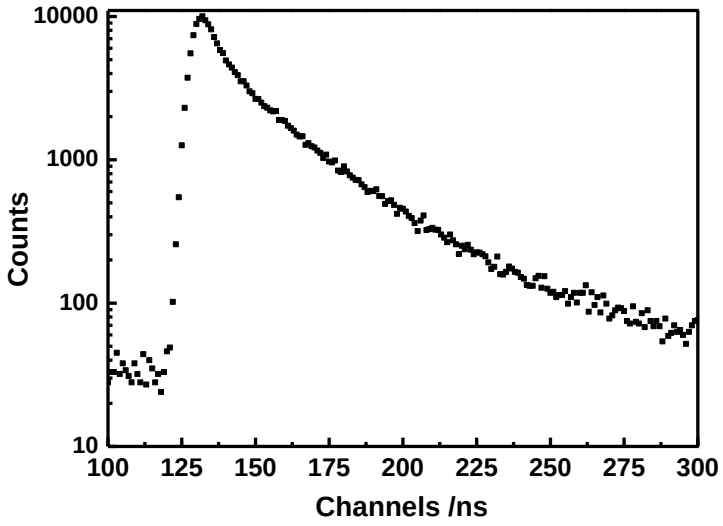


Fig.S6 The photograph of the products from different molar ratios of CA and DCD under 365 nm UV light.

On the upper row, the products were synthesized by tuning the amount of dicyandiamide. 0.21 g (1 mmol) of citric acid was separately reacted with 0.007 g (0.083 mmol), 0.009 g (0.11 mmol), 0.015 g (0.15 mmol), 0.028 g (0.33 mmol), of dicyandiamide, making the molar ratio of citric acid to dicyandiamide vary from 12:1 to 9:1, 6:1, and 3:1. Oppositely, in the next row, the products were synthesized by tuning the amount of citric acid. 0.25 g (3 mmol) of dicyandiamide was separately reacted with 0.63 g (3 mmol), 0.21 g (1 mmol), 0.11 g (0.5mmol), 0.07 g (0.33 mmol) and 0.05g (0.25mmol) of dicyandiamide, making the molar ratios of dicyandiamide to citric acid vary from 1:1 to 3:1 , 6:1, 9:1, and 12:1.



τ_i / ns	A_i / %
3.86	**45.73**
9.31	**30.63**
0.73	**23.64**

Fig.S7 Fluorescence lifetime intensity decay of GQDs in aqueous solution (excitation at 370 nm, emission at 452 nm).

According to Equation (1)

$$\bar{\tau} = \frac{A_1\tau_1 + A_2\tau_2 + A_3\tau_3}{A_1 + A_2 + A_3} \quad (1)$$

the average fluorescence lifetime of GQDs was calculated as 4.78 ns, wherein A_i is the fractional contributions of time-resolved decay lifetime of τ_i.

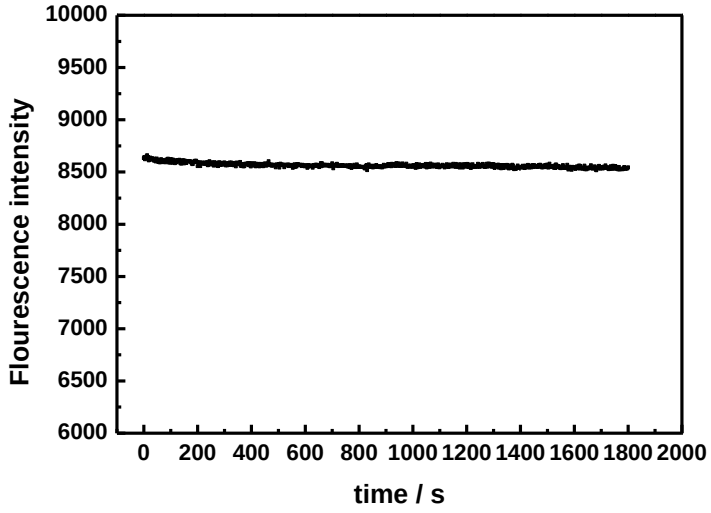


Fig.S8 Effect of time on the fluorescence intensity of the GQDs.

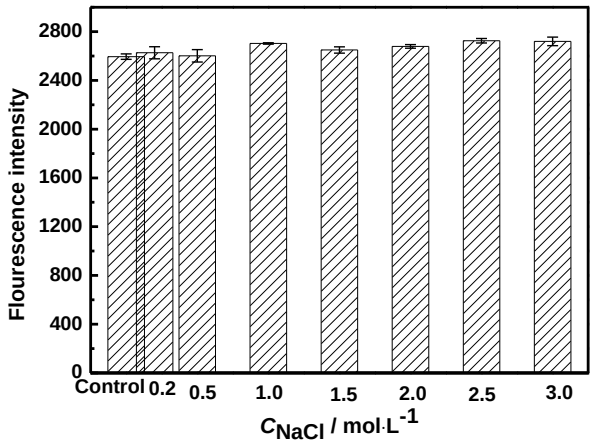


Fig.S9 Effect of ionic strengths on the fluorescence intensity of GQDs (ionic strengths were controlled by various concentrations of NaCl)

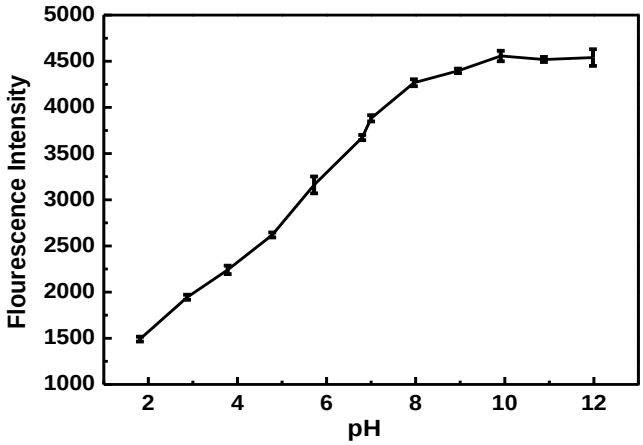


Fig.S10 The fluorescence intensity of the GQDs varied with different pH.

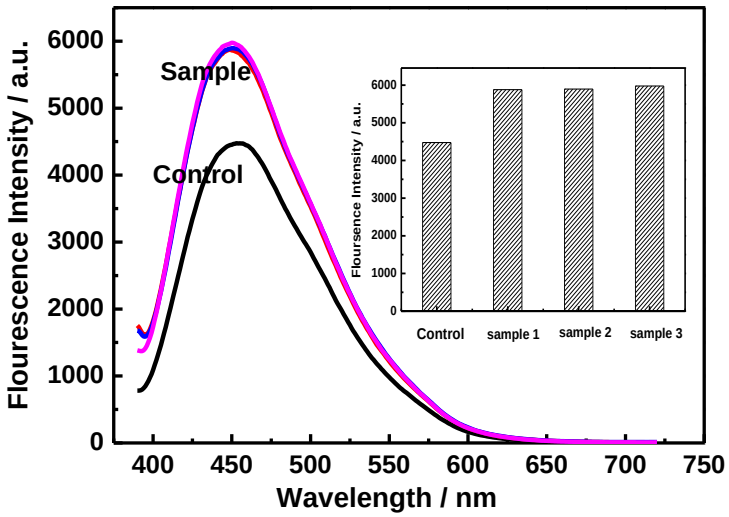


Fig.S11 The fluorescence intensity of CDs changes in serum.

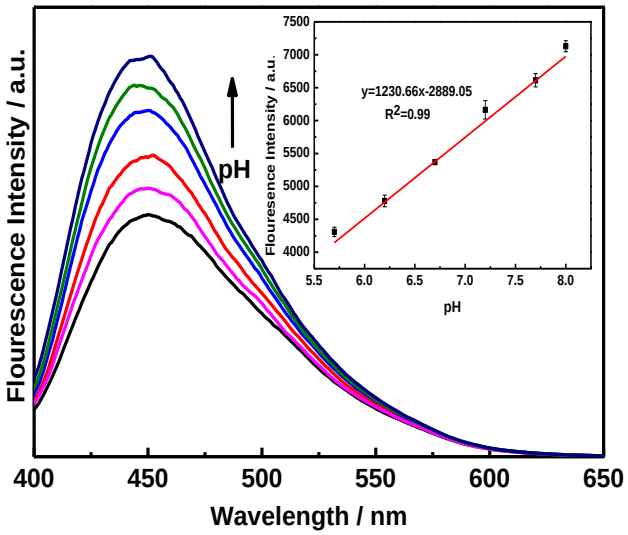


Fig.S12 The fluorescence spectra of GQDs in different pH PBS buffer (5.7, 6.2, 6.7, 7.2, 7.8, and 8.0). Inset: the linear relationship between fluorescence intensity and pH.

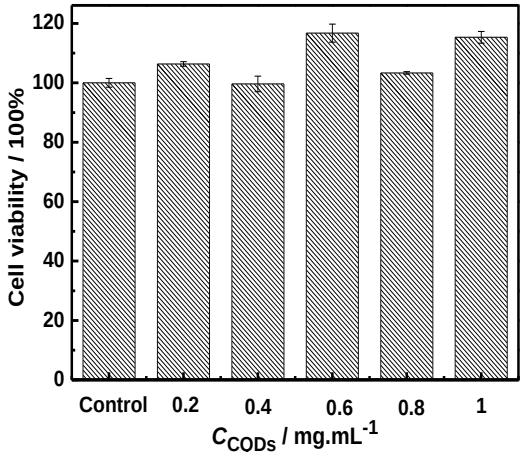


Fig.S13 The Cellular cytotoxicity of the synthetized GQDs.