Supporting information

Polyimide-pyrolyzed Carbon Wastes Approach to Scalable and Controlled Electrochemical Preparation of Size-Tunable Graphene

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EXPERIMENTAL SECTION

Yield of af-GQDs. The yield of GQDs is calculated as follows:

$$\text{yield}=\frac{M_{GQDs}}{M_r}\times 100\%$$

Where M_{GQDs} is the mass of the obtained GQDs, and M_r is the parts of the materials that take part in the electrochemical reaction.

C conversion of GQDs. The C conversion of GQDs calculated as follows:

C conversion=C mass of GQDs/C mass of the materials.

Where C mass of GQDs are obtained by the M_{GQDs}×percentage composition of C, and C mass of materials are obtained by the M_r × percentage composition of C. the percentage composition are obtained from the XPS results.

Concentration of GQDs. It was calculated by the equation:

$$C_{GQD}=\frac{W_{GQD}}{L}$$

In this equation, the C_{GQD} is the concentration of GQDs. W_{GQD} is the mass of GQDs, L is the volume of the solution after the electrochemical reaction.

Quantum Yield (φ) of af-GQDs. The quantum yield of GQDs calculated as follows:

$$\varphi=\varphi_R\frac{I}{I_R}\frac{A}{A_R}\frac{\eta^2}{\eta_R^2}$$

Where I is the measured integrated emission intensity, η is the refractive index of the solvent, A refers to the optical density, and the subscript R denotes the reference standard with a known φ (quinine sulfate in sulfuric acid solution, φ_R = 0.54).

Electrochemical calculation. Specific capacitance of the 3DGCs was caculated from GCD discharge curve:

$$C_s=\frac{2\times I\times t}{m\times\Delta U}$$

Where C_s (F/g), I (A), t (s), m (g) and ΔU (V) are the gram specific capacitance, the discharge current, the discharge time, the mass of single electrode and the potential window, respectively.

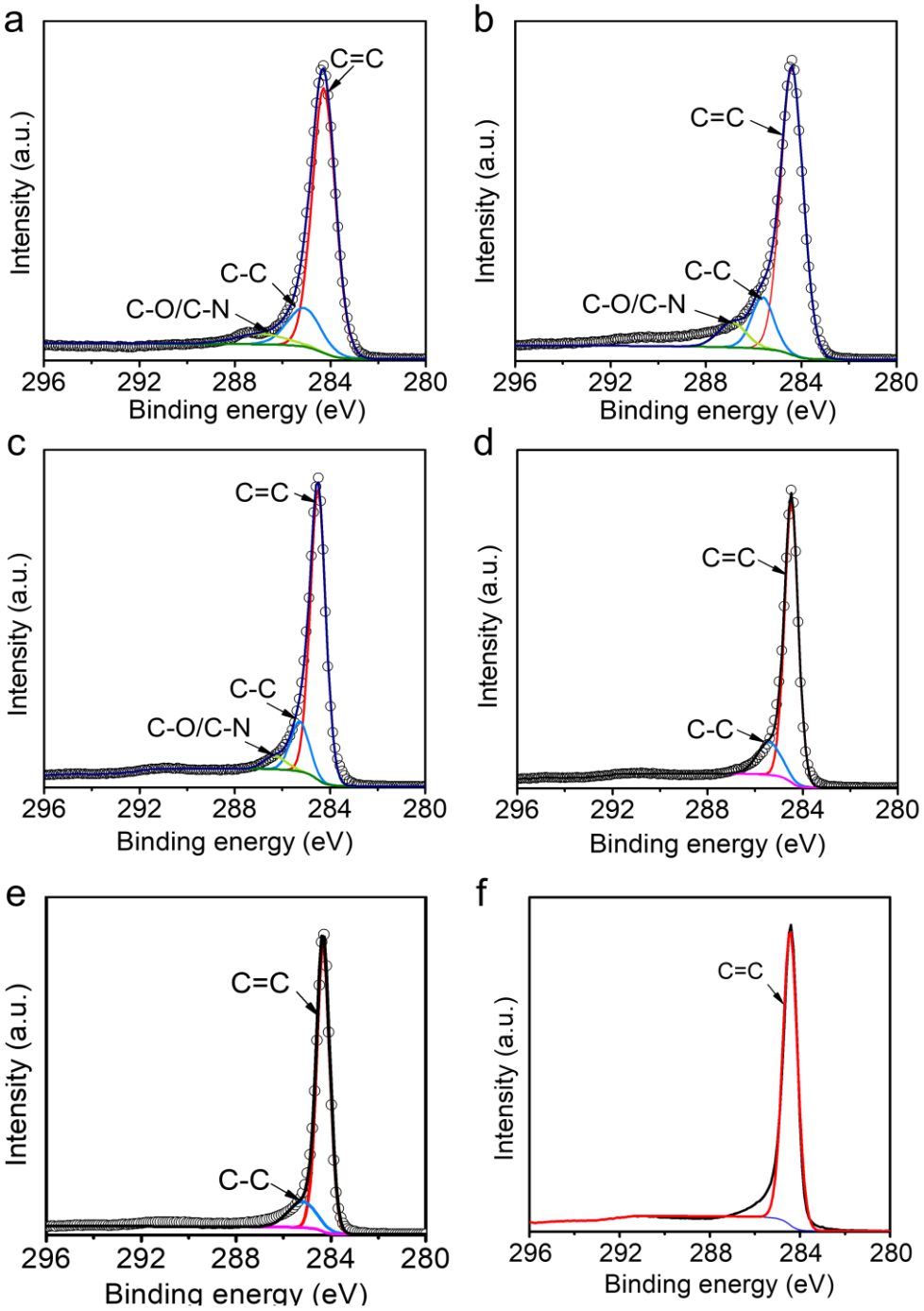

Figure S1. (a-f) The C 1s spectra of the PPC-800, PPC-1300, PPC-1600, PPC-2000, PPC-2300 and PPC-2800, respectively.

Table S1. Comparison of the C_r of different PPC films.

Materials	PPC-800	PPC-1300	PPC-1600	PPC-2000	PPC-2300	PPC-2800
r of sp^2 C	66.7%	78.1%	78.7%	82.6%	92.7%	100%
r of sp^3 C	33.3%	21.9%	21.3%	17.4%	7.3%	0
C_r	0.5	0.28	0.27	0.21	0.08	0

r is the proportion of sp^x C.

Table S2. The C, O, N contents of PPC films.

	C（%）	O（%）	N（%）
PPC-800	93.6	3.1	3.3
PPC-1300	98.1	1.2	0.74
PPC-1800	97.5	2.5	-
PPC-2300	98	2	-
PPC-2800	100	0	-

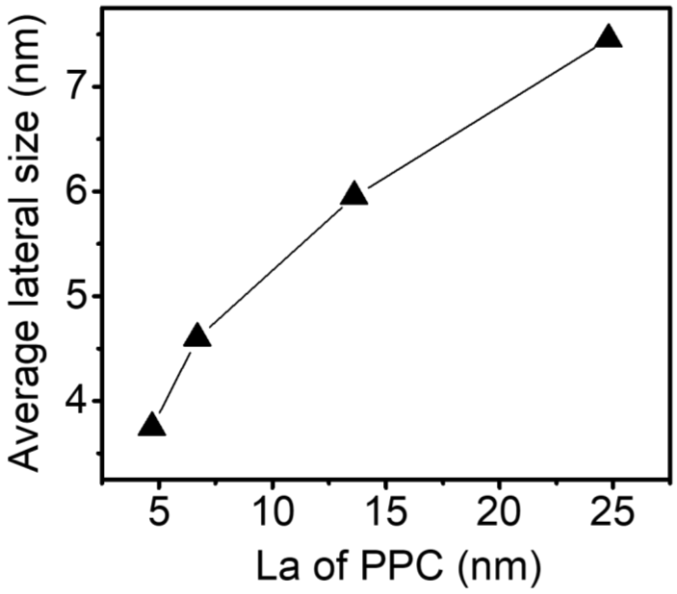

Figure S2. The changes of average lateral size of GQDs with La of PPC.

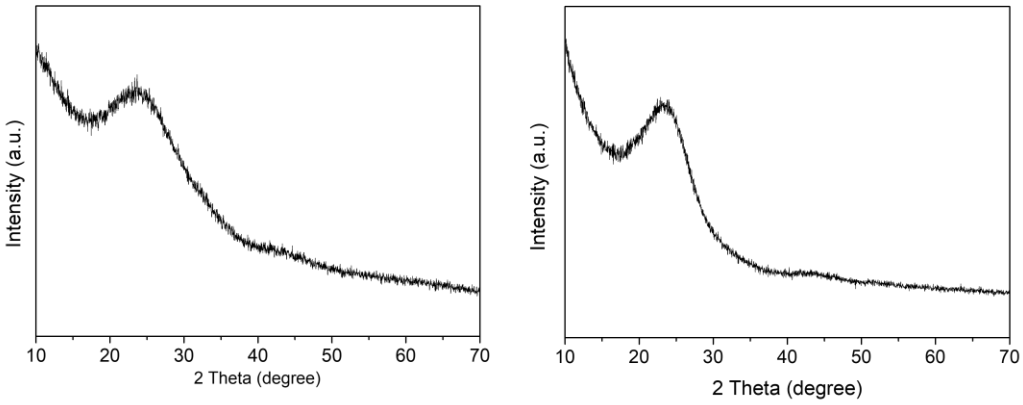

Figure S3. XRD patterns of PPC-800 and PPC-1300.

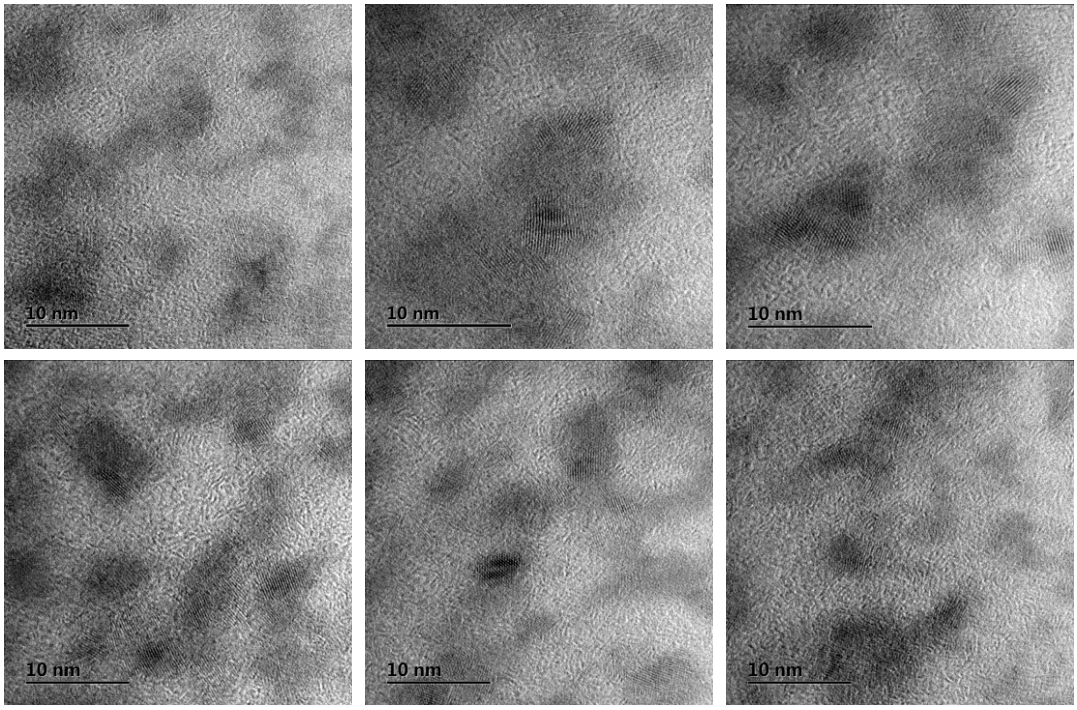

Figure S4. The typical TEM images of GQDs-800.

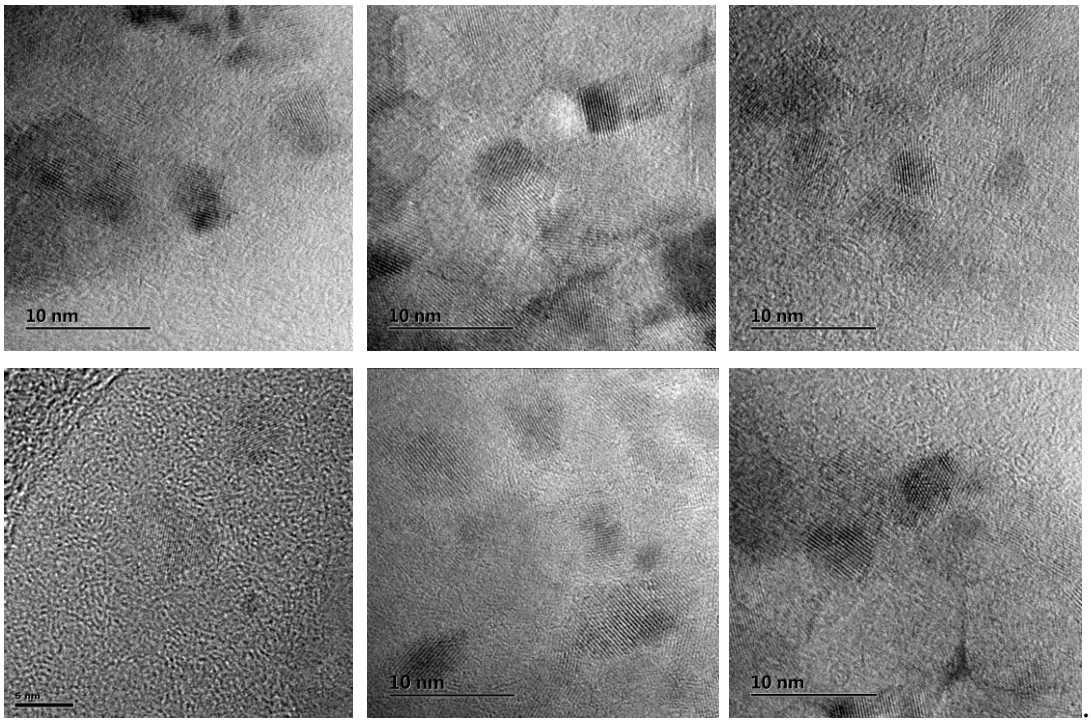

Figure S5. The typical TEM images of GQDs-1300.

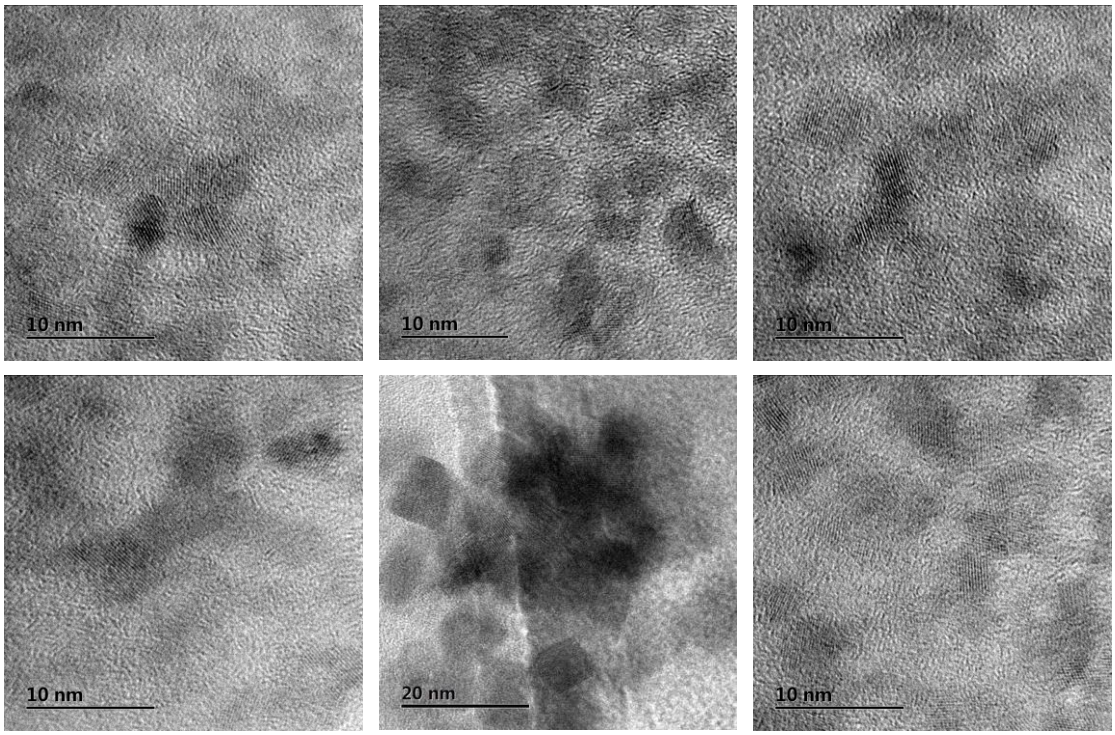

Figure S6. The typical TEM images of GQDs-1600.

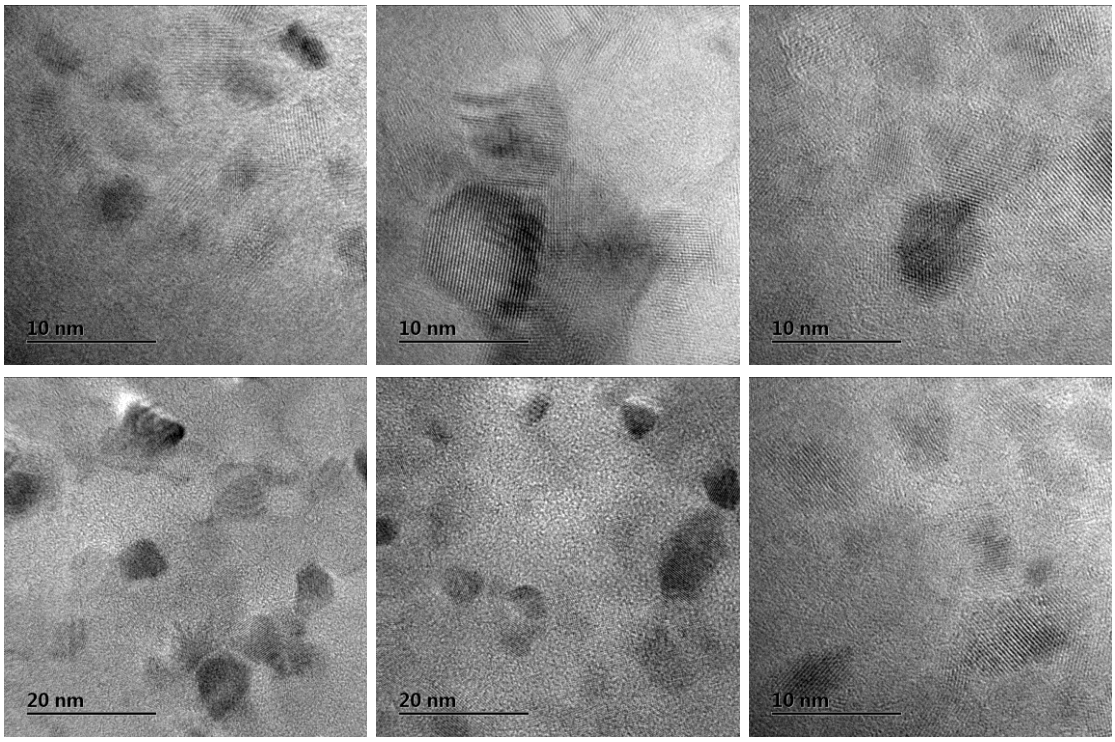

Figure S7. The typical TEM images of GQDs-2000.

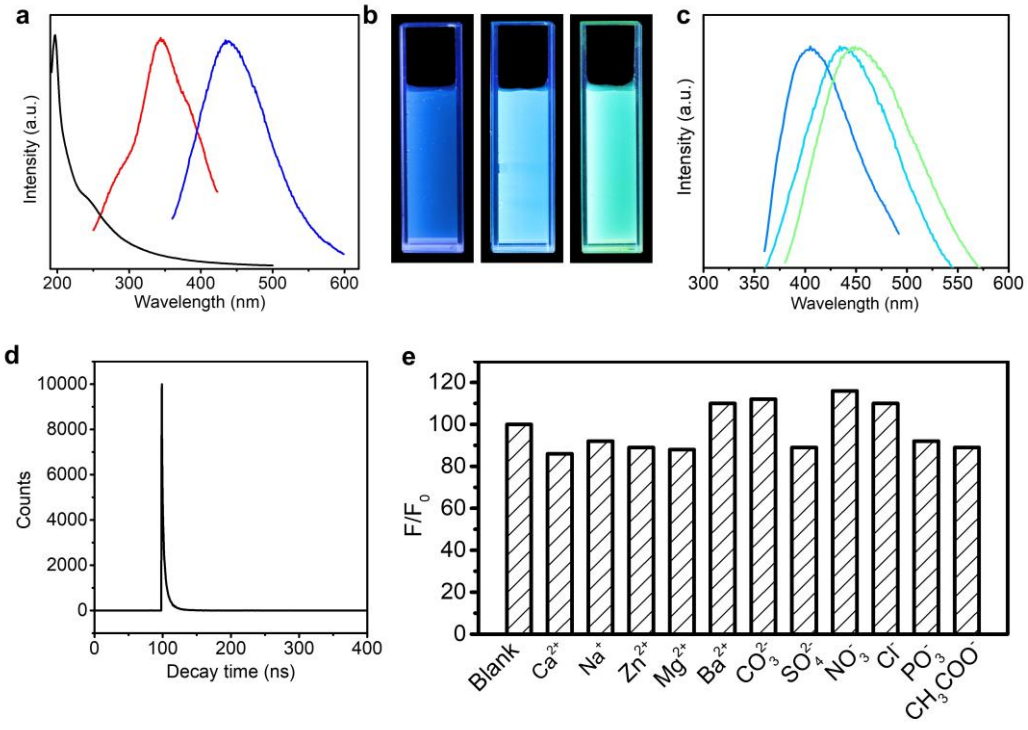

Figure S8. (a) UV−vis absorption (black curve), PL (blue curve), and PL excitation (red curve) spectra of the GQDs. (b) fluorescence shift under UV light. (c) Their typical

PL spectra (d) Time-resolved PL spectrum. (e) Dependence of PL intensity on ions. F_0 and F are PL intensities in the absence and presence of ions. The concentration of the ions is 0.05 M.

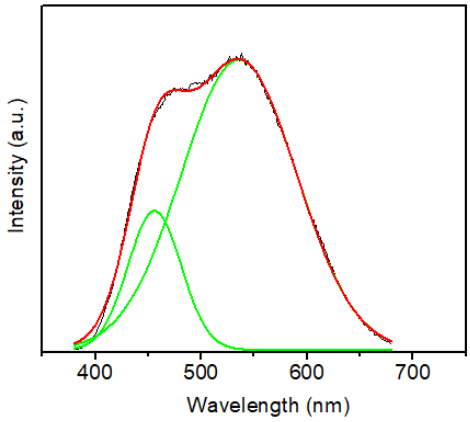

Figure S9. The PL spectrum of GQDs-2300 prepared by using the mixture electrolyte of ammonium hydroxide and terephthalic acid.

Optical properties are the significant factors to value the GQDs. Using GQDs-1300 with high crystallinity as a model. The ultraviolet visible (UV−vis) absorption spectrum (Figure S8a) shows a typical π-π* transition of the aromatic sp^2 domains at 198 nm and n-π* transition absorption peak at 247 nm. Their water dispersion exhibits excitation dependent PL with emission maximum at 438 nm, which is excited by the wavelength of 343 nm. Notably, the fluorescence shows red shift by increasing graphitization degree of PPC (Figure S8b-c). The maximum emission wavelength ranges from 405 nm to 453 nm. The more certain phenomenon is observed at the mixture electrolyte of ammonium hydroxide and p-phthalic acid (PTA). Because the PTA can give effective coverage on PPC anode, resulting in better crystallinity of these GQDs. Moreover, adding of strong acid can etch PPC films with high graphitization (annealing after 2300 ^{o}C-2800 ^{o}C) and acquire the yellow fluorescence (Figure S9). The QY enhances (from 11.2% to 28%) by virtue of the good crystallinity. The PL decay is monoexponential with lifetime of 3.05 ns (Figure S8d), suggesting intrinsic PL characteristic of GQDs-1300. The ions response well reflects the PL stability at various saline solution, giving an application prospect in bio-correlated fields (Figure S8e).

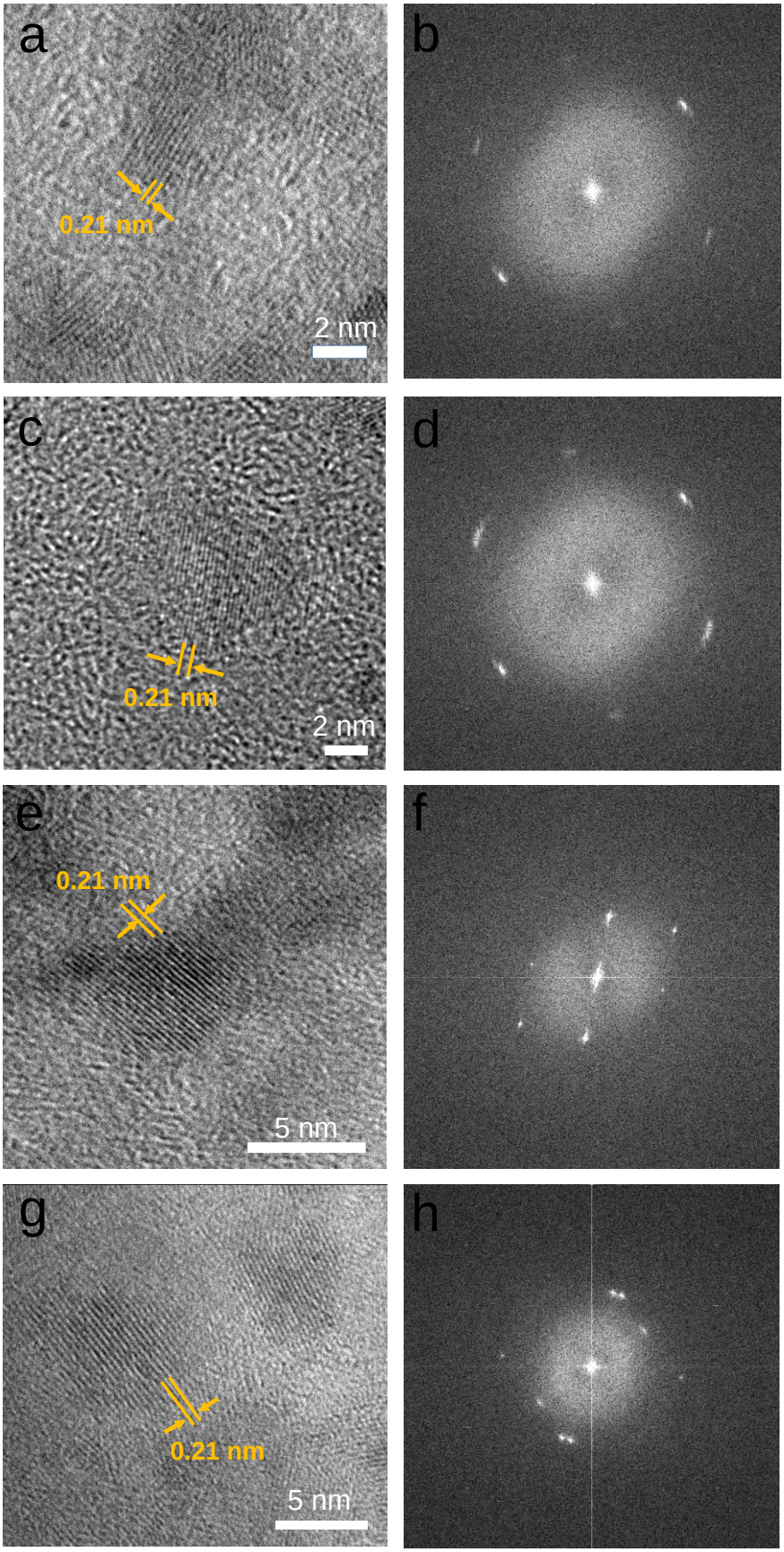

Figure S10. HRTEM image and FFT pattern of (a, b) GQDs-800, (c, d) GQDs-1300, (e, f) GQDs-1600, (g, h) GQDs-2000.

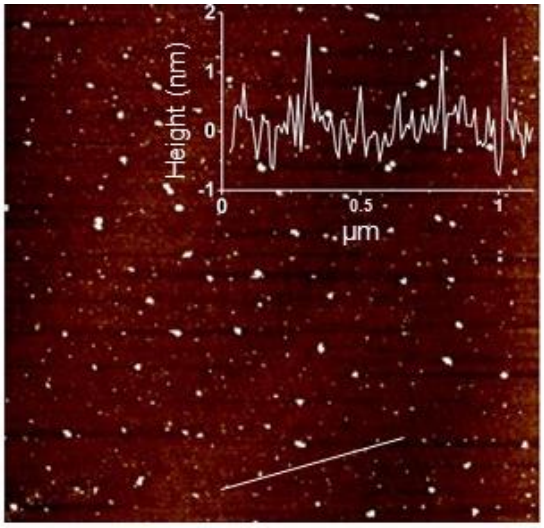

Figure S11. AFM image of the GQDs-1300.

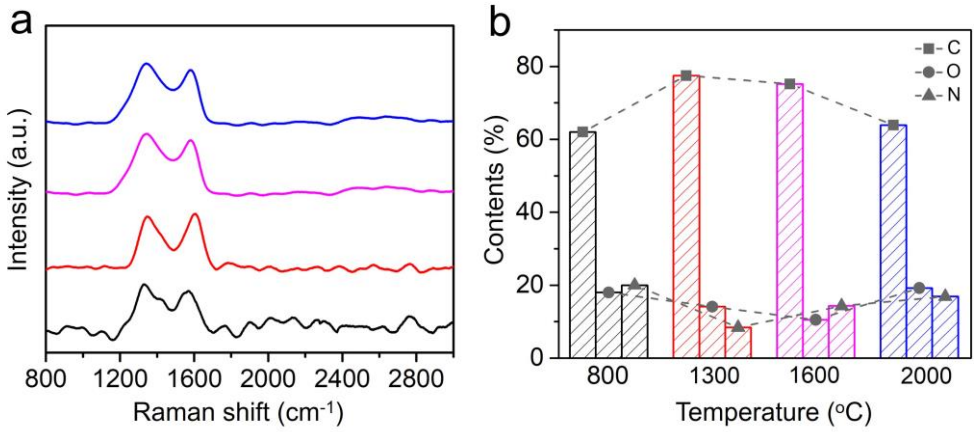

Figure S12. (a) Raman spectra and (b) the C, O, N contents the GQDs-800 (black mark), GQDs-1300 (red mark), GQDs-1600 (pink mark) and GQDs-2000 (blue mark).

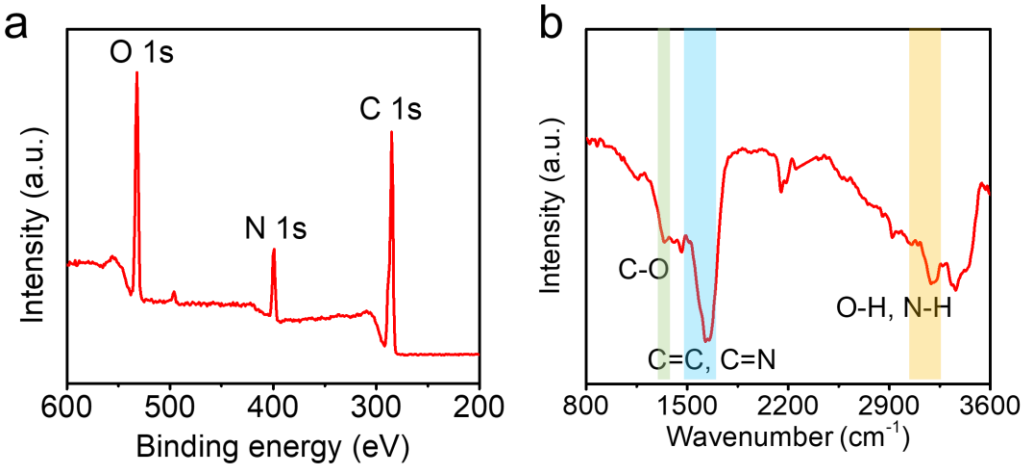

Figure S13. (a) XPS survey and (b) FT-IR spectrum of GQDs-1300.

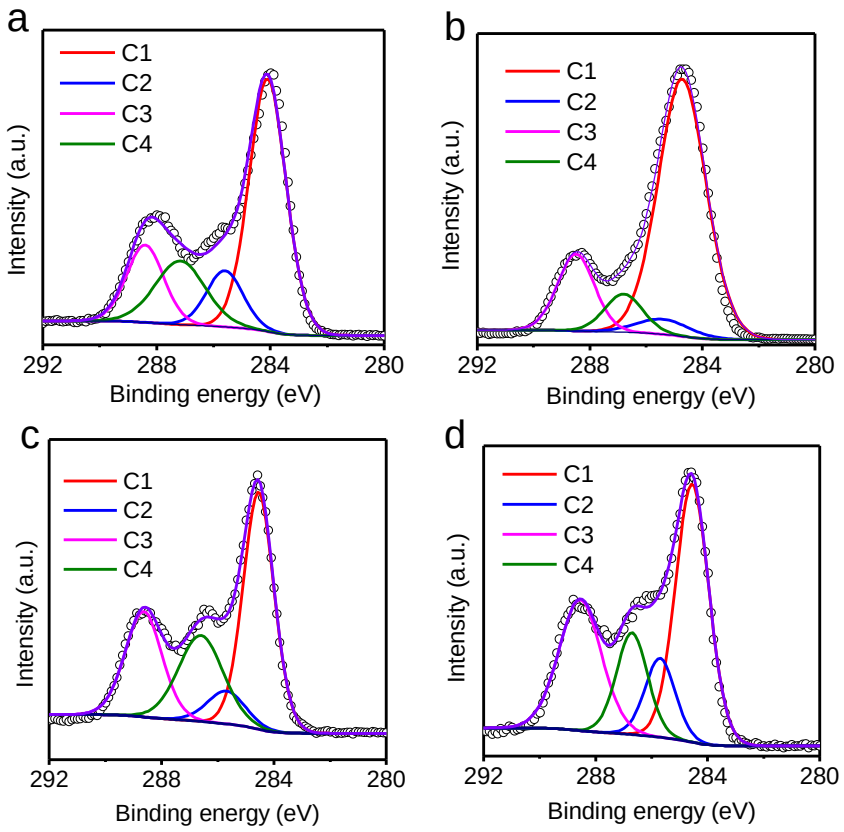

Figure S14. The C 1s spectra of (a) GQDs-800, (b) GQDs-1300, (c) GQDs-1600 and (d) GQDs-2000, respectively. The C=C (284.5 eV), C–C, C–H and C–N (285.3 eV), O–C=O (286.7 eV), and C=O (288.5 eV) are shown as C1, C2, C3 and C4.

The Raman spectrum (Figure S12a) of GQDs shows the typical D peak and G peak with an I_D/I_G intensity ratio of 2±0.3, suggesting sufficient oxidation of PPC before cut into the electrolyte. However, the oxidation degree of the GQDs depends on the structure of the PPC. The detailed element content illustrated by XPS surveys (Figure S12b). GQDs-800 exhibit only 62.1% of C due to high percentage and easy-to-oxidized sp^3 C. About 77.5% of C is contained in the GQDs-1300, ascribing to the superiority of PPC-1300 with bicontinuous structure and suitable C_r. Furthermore, the C=N and N-H bond are found in the GQDs, suggesting the functionalized feature of these GQDs (Figure S13-14).

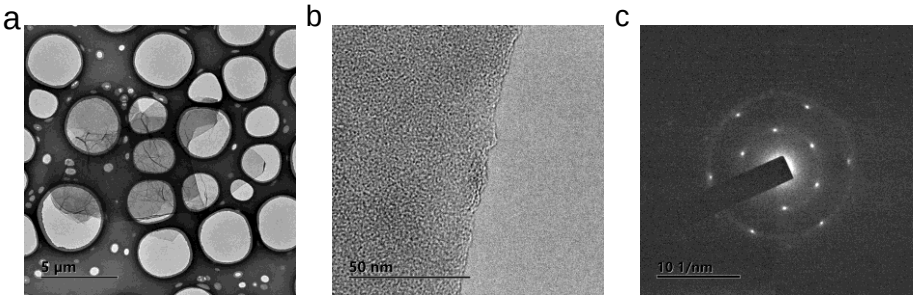

Figure S15. (a) TEM, (b) HRTEM images and (c) SAED pattern of graphene prepared by PPC-2300.

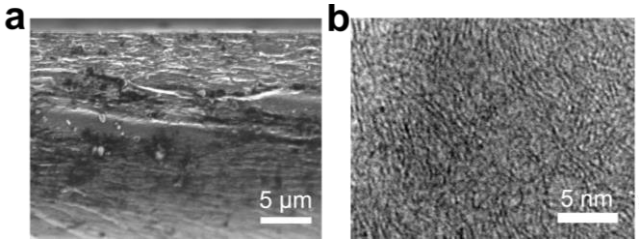

Figure S16. (a) SEM image of the edge of fractured PPC-1600, (b) HRTEM image of the surface of PPC-1600.

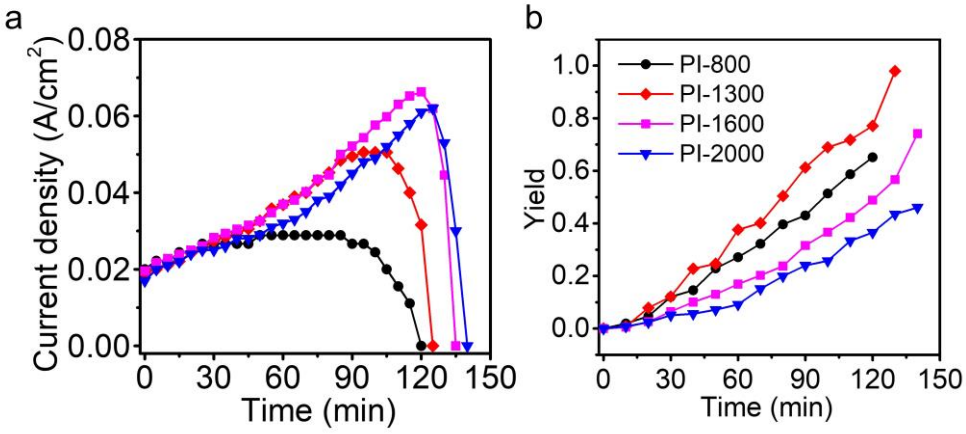

Figure S17. Current density-Time Curve and (i) Yield-Time Curve of GQDs-800 (black mark), GQDs-1300 (red mark), GQDs-1600 (pink mark) and GQDs-2000 (blue mark)

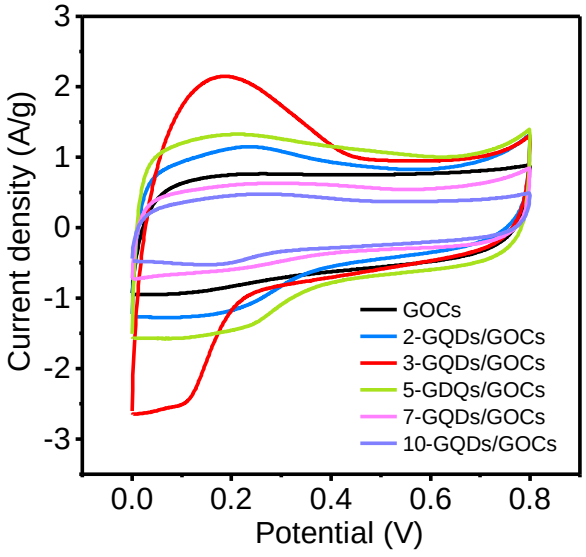

Figure S18. CV tests at scan rate of 10 mV/s.

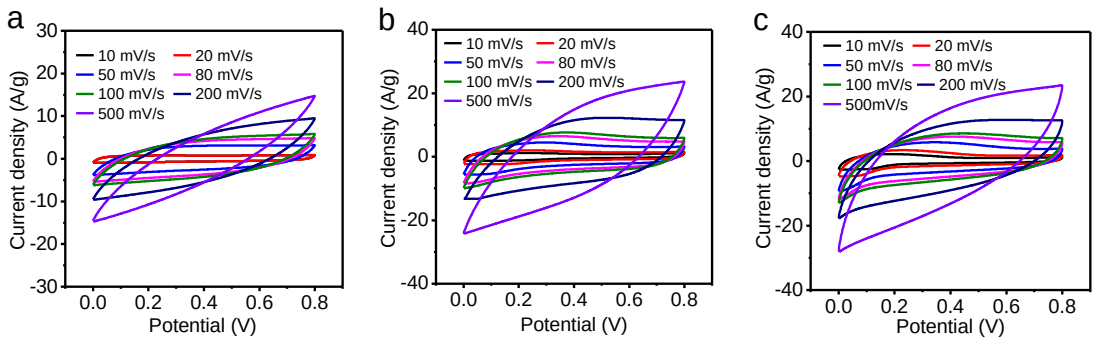

Figure S19. CV curves for (a) GOCs, (b) 2-GQDs/GOCs, (c) 3-GQDs/GOCs at scan rates from 10 to 500 mV/s.

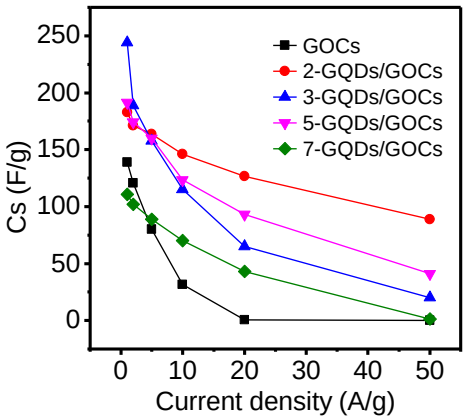

Figure S20. Rate performance while current density increased from 0.1 A/g to 50 A/g.