Graphene-Coupled Nitrogen-Enriched Porous Carbon Nanosheets for Energy Storage

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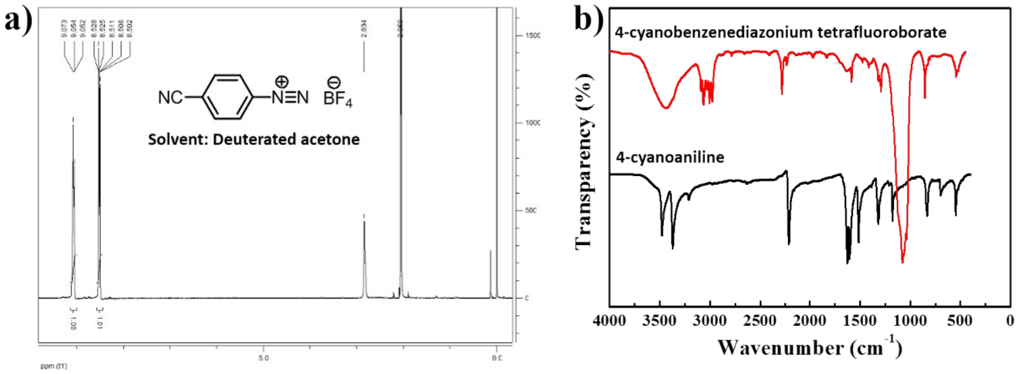

Fig. S1 a) ^{1}H NMR spectrum of 4-cyanobenzenediazonium tetrafluoroborate, b) FTIR spectra of 4-cyanoaniline and 4-cyanobenzenediazonium tetrafluoroborate.

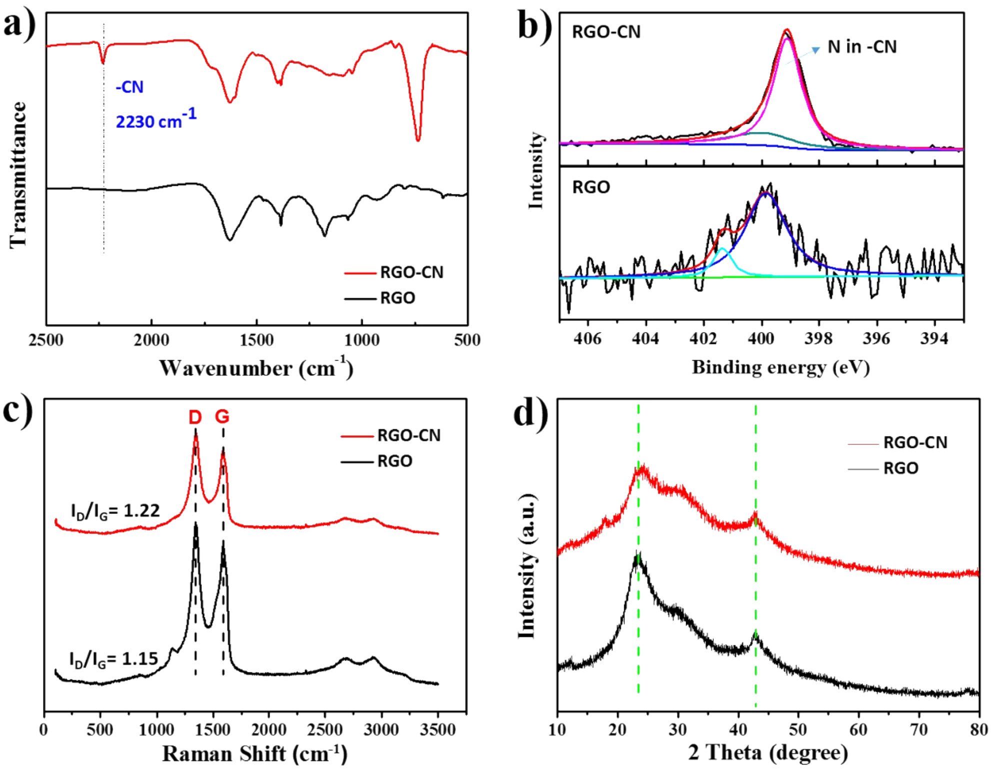

Fig. S2 Structure analysis for RGO and RGO-CN, a) FTIR spectra, b) N1s XPS spectra, c) Raman spectra and d) XRD patterns.

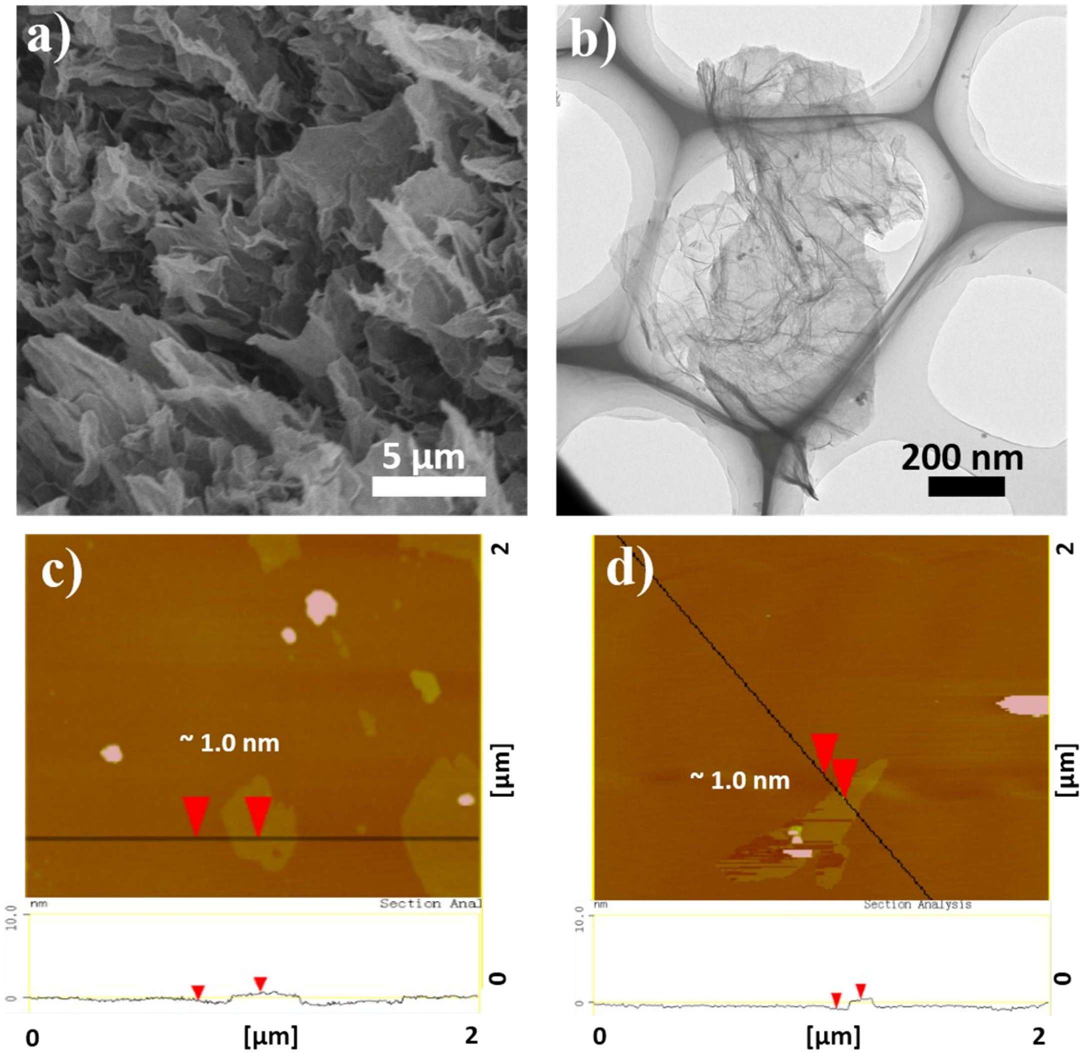

Fig. S3 Morphology and microstructure of RGO-CN, a) SEM, b) TEM and c) AFM images. AFM image of RGO (d).

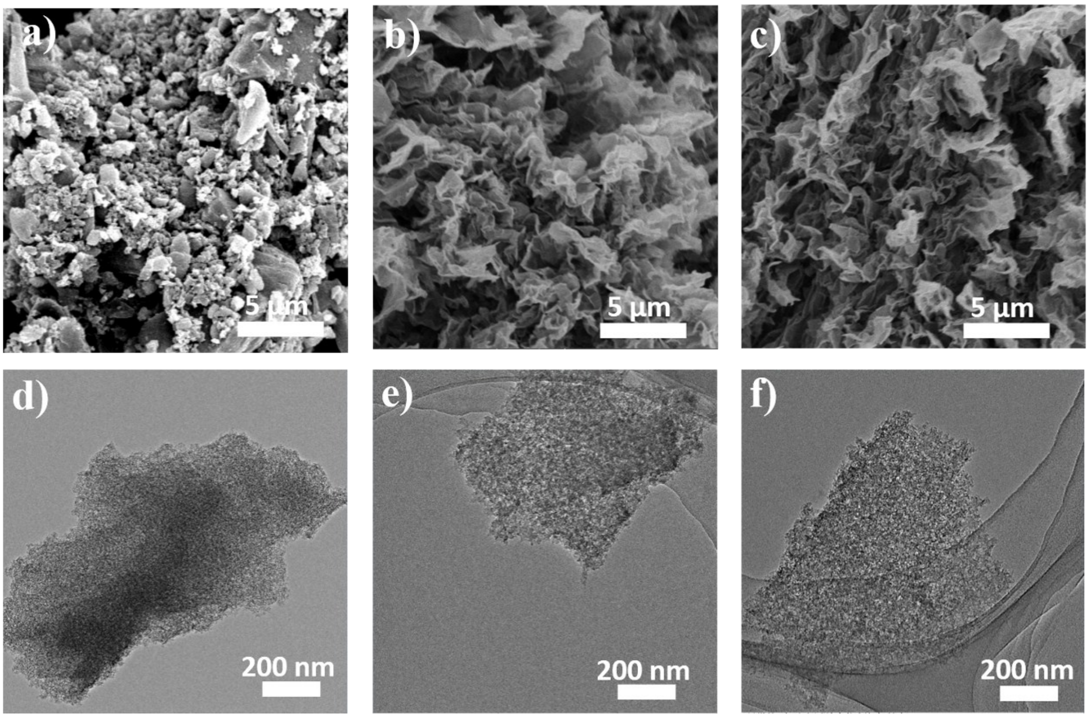

Fig. S4 SEM images of CTF (a), G-CTF-10 (b) and G-CTF-20 (c); TEM images of CTF (d), G-CTF-10 (e) and G-CTF-20 (f).

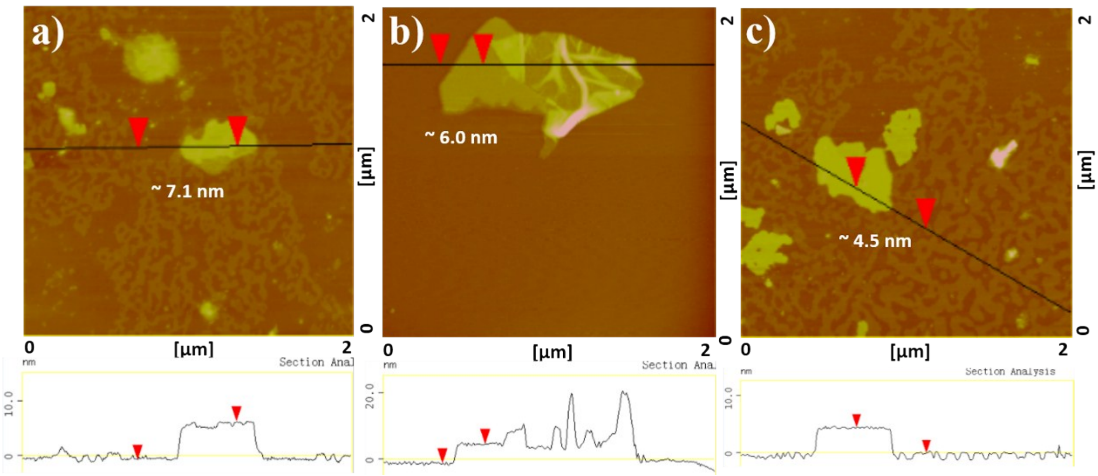

Fig. S5 AFM images and height profiles of G-CTF-10 (a), G-CTF-15 (b) and G-CTF-20 (c).

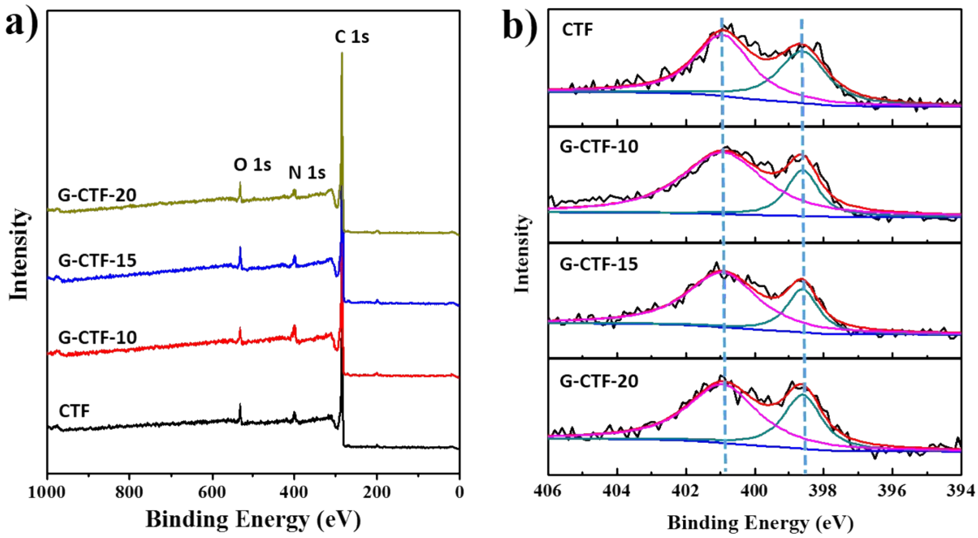

Fig. S6 XPS survey spectra (a) and N1s XPS spectra (b) of CTF and G-CTFs.

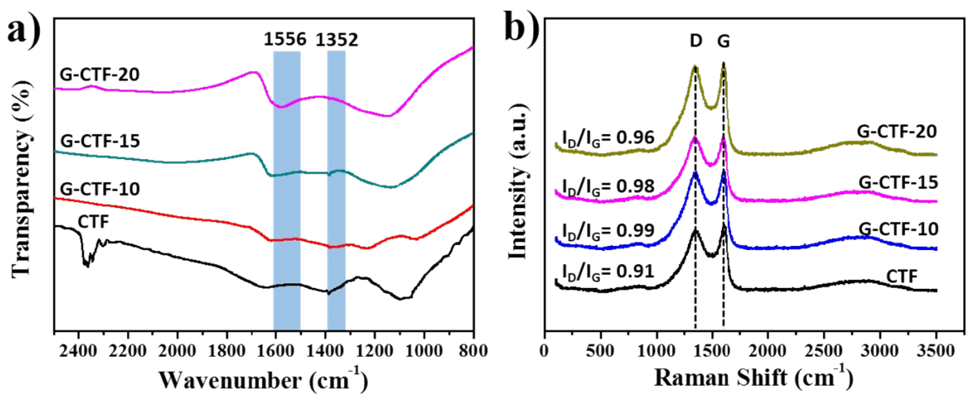

Fig. S7 FTIR spectra (a) and Raman spectra (b) of CTF and G-CTFs.

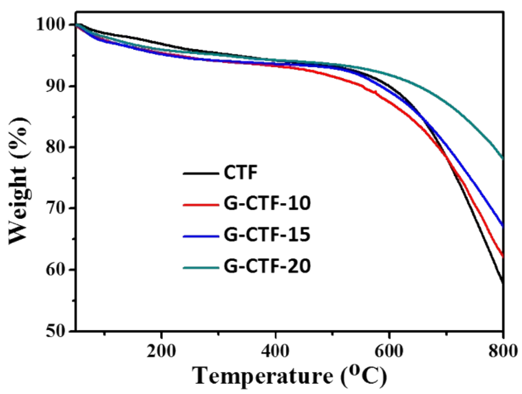

Fig. S8 TGA curves of CTF and G-CTFs.

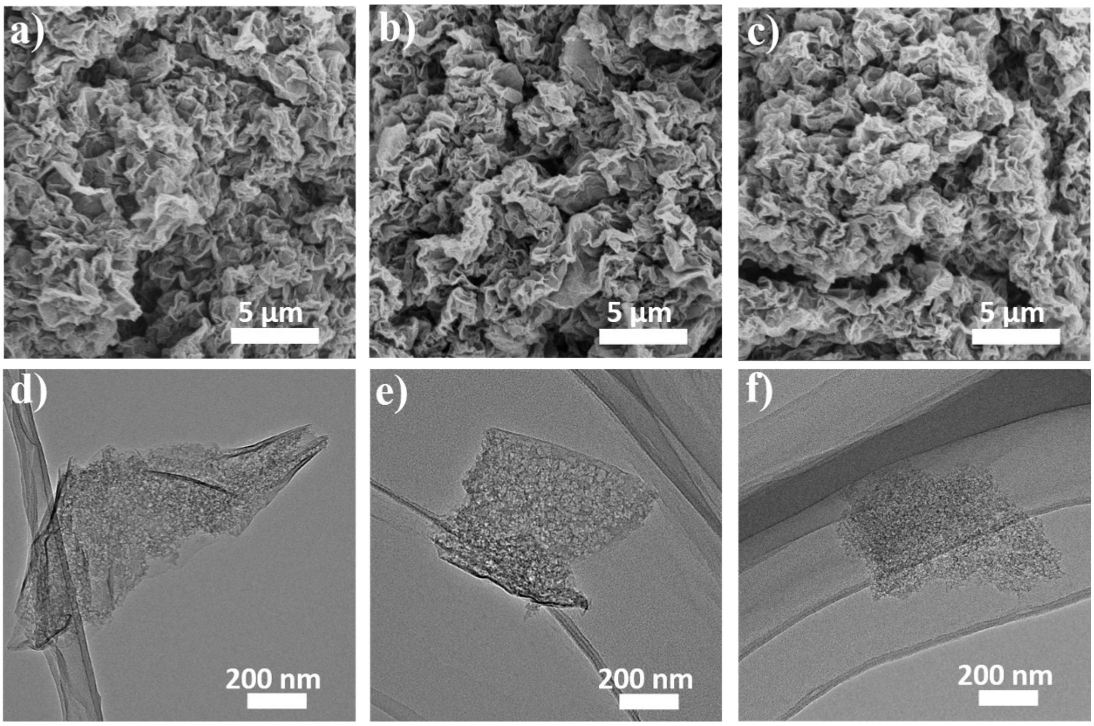

Fig. S9 SEM images of G-PC-700 (a), G-PC-800 (b) and G-PC-900 (c); TEM images of G-PC-700 (d), G-PC-800 (e) and G-PC-900 (f).

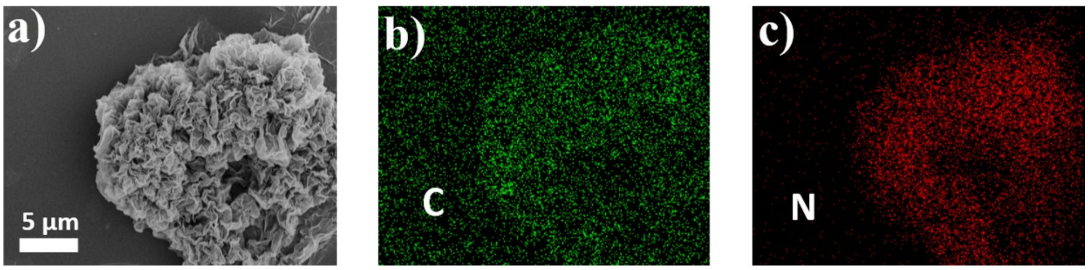

Fig. S10 SEM images of G-PC-800 (a) and the corresponding elemental mapping images (b and c).

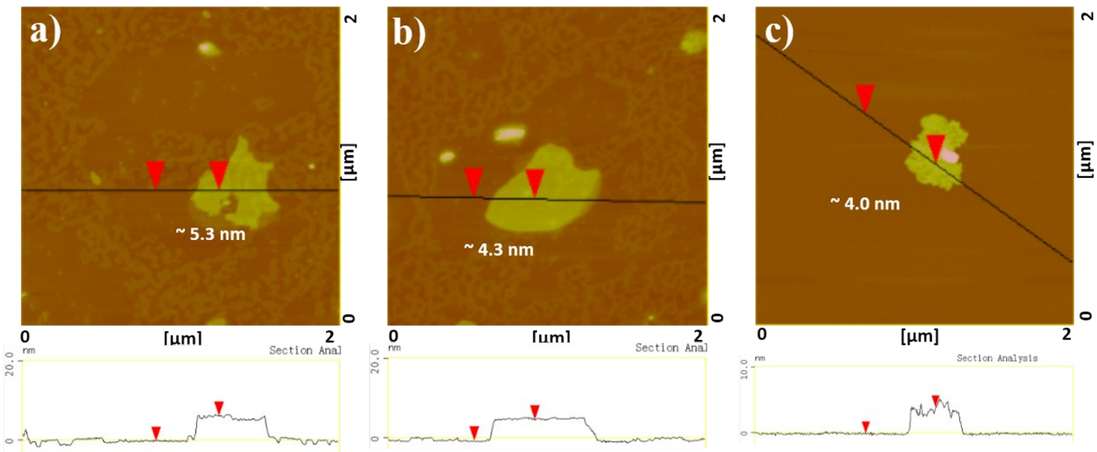

Fig. S11 AFM images and height profiles of G-PC-700 (a), G-PC-800 (b) and G-PC-900 (c).

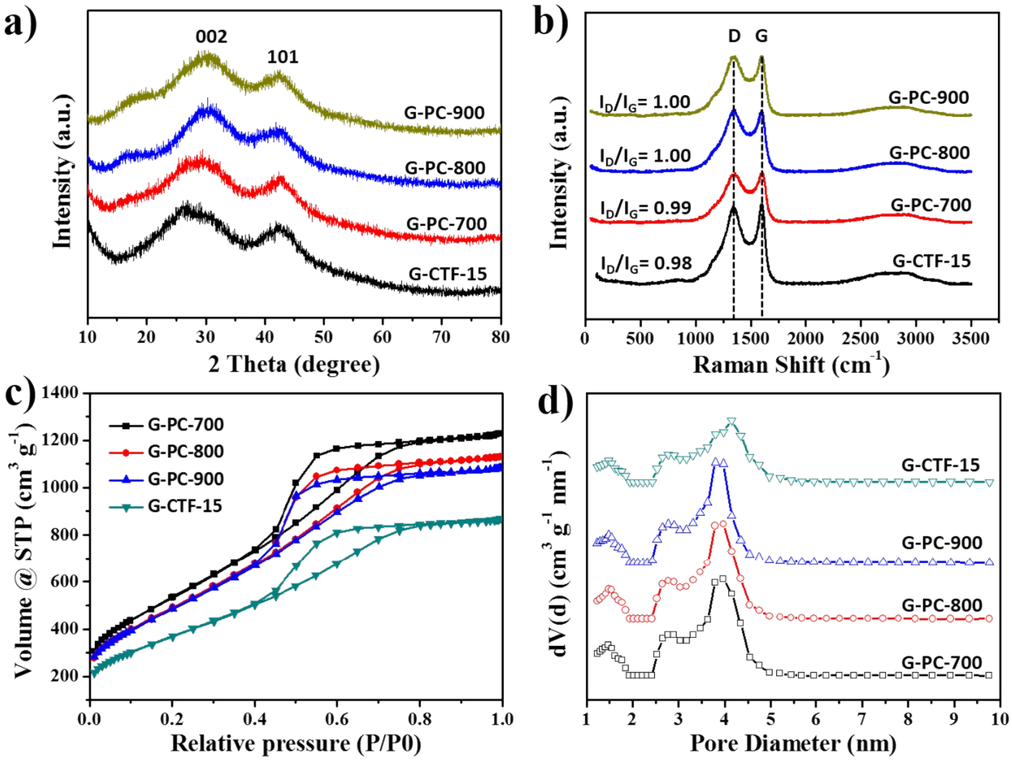

Fig. S12 Structure analysis for G-CTF-15 and G-PCs. (a) XRD patterns, (b) Raman spectra, (c) nitrogen adsorption/desorption isotherms and (d) the corresponding pore size distribution.

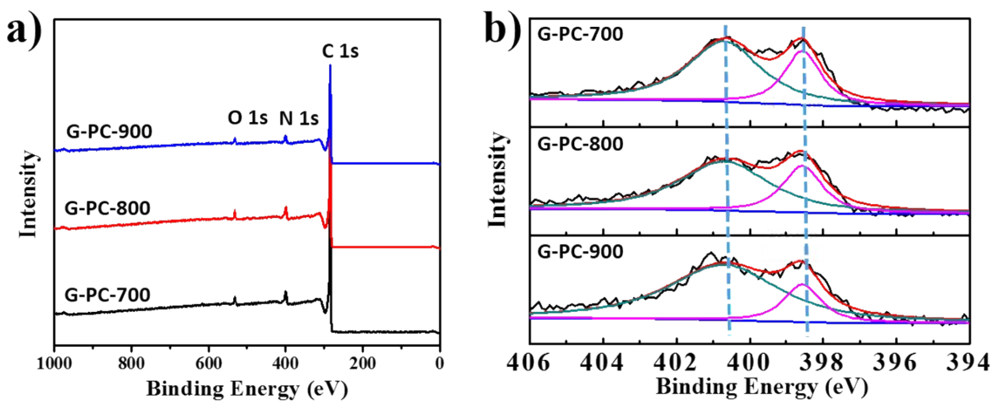

Fig. S13 XPS survey spectra (a) and N1s XPS spectra (b) of G-PCs.

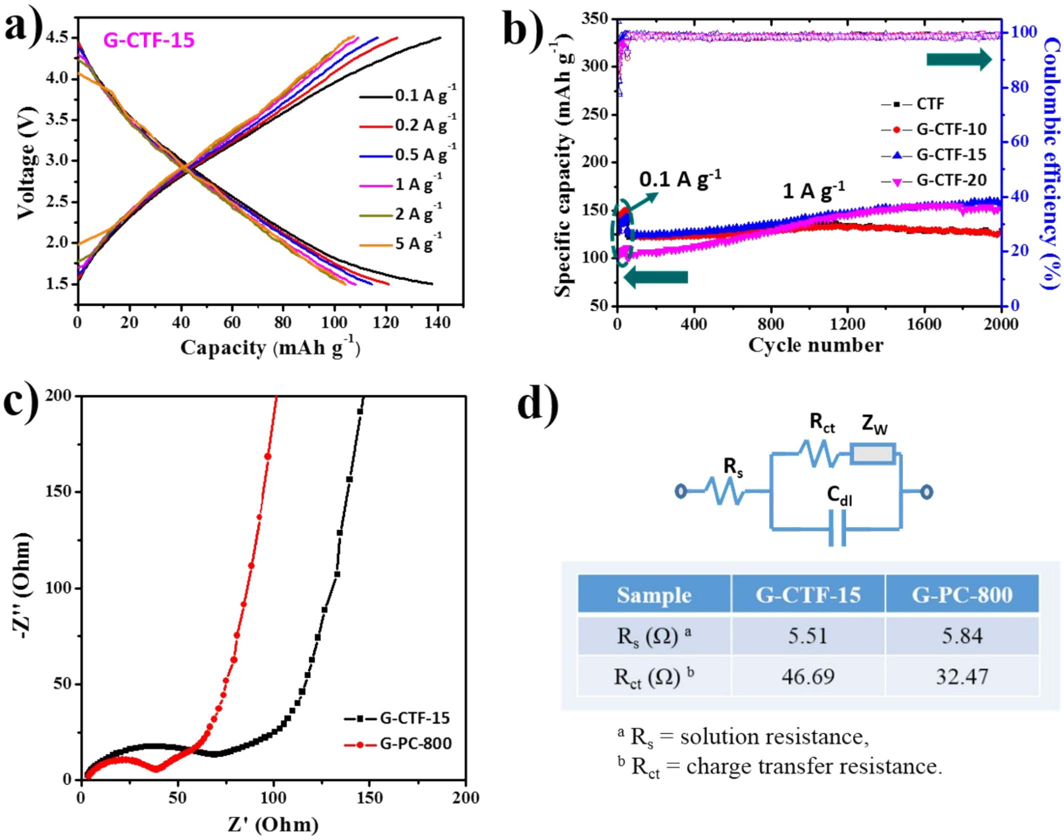

Fig. S14 The electrochemical performance of G-CTFs|Li half cells in the voltage range of 1.5-4.5 V *versus* Li/Li^+. a) Charge-discharge curves of G-CTF-15 electrode with varying current densities. (b) Cycle performance of CTF and G-CTFs electrodes with a current density of 0.1 A g^{-1} for the first 50 cycles and 1 A g^{-1} for the consequent cycles. Nyquist plots (c), equivalent circuit and kinetic parameters (d) of G-CTF-15 and G-PC-800.

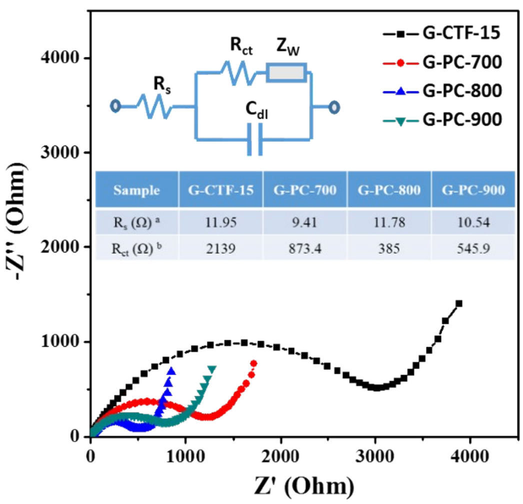

Fig. S15 Nyquist plots of G-CTF-15 and G-PCs. Inset: equivalent circuit. R_s, R_{ct}, C_{dl} and Z_W represent the electrolyte resistance, the charge transfer resistance, the double layer capacitance and the

Warburg impedance, respectively.

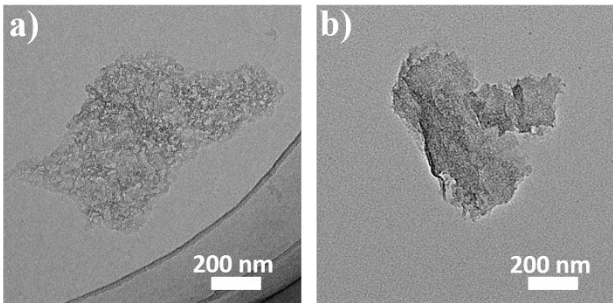

Fig. S16 TEM images of G-PC-800 after 3000 cycles and 500 cycles in Li-ion battery (a) and Na-ion battery (b), respectively.

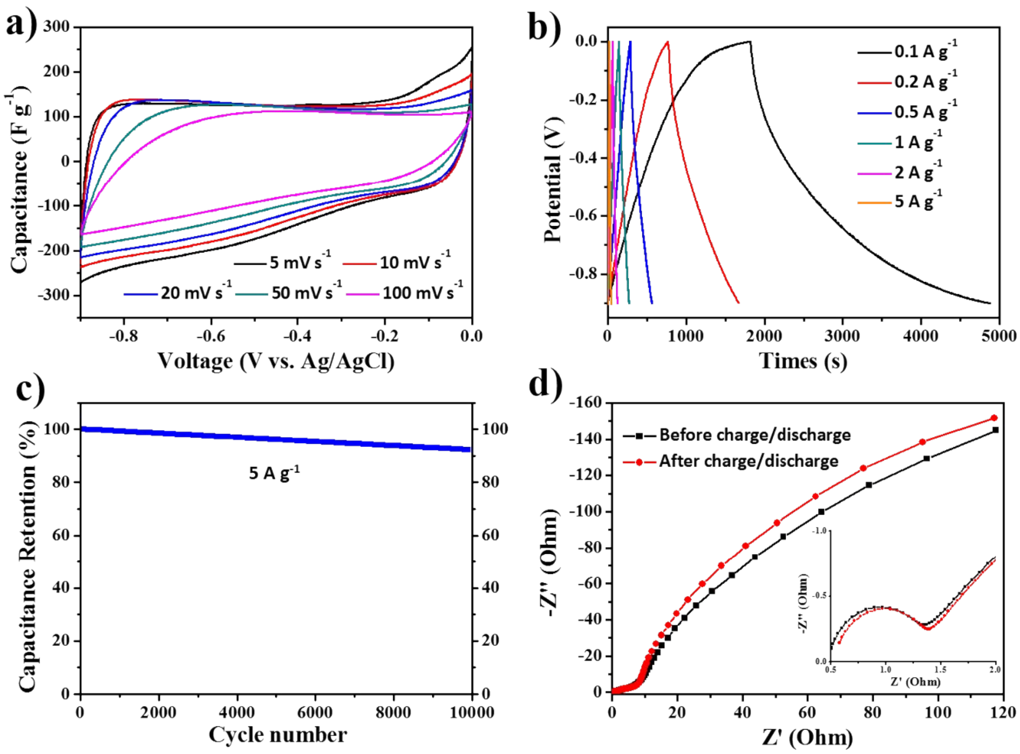

Fig. S17 Capacitive performance of G-PC-800 electrode. a) CV curves for different scan rates in 6 M KOH, b) galvanostatic charge-discharge curves at different current densities in 6 M KOH, c) cycle performance at 5 A g^{-1} for 10,000 cycles, and d) Nyquist plots before and after charge and discharge under open-circuit voltage.

Table S1. Nitrogen contents of CTF, G-CTFs and G-PCs from Elemental Analysis (EA) and corresponding nitrogen species concentration calculated by XPS.

Sample	CTF	G-CTF-10	G-CTF-15	G-CTF-20	G-PC-700	G-PC-800	G-PC-900
N content (wt%) [a]	**8.67**	**7.34**	**6.10**	**5.92**	**7.05**	**6.35**	**5.49**
N/C [b]	**0.10**	**0.15**	**0.10**	**0.08**	**0.13**	**0.12**	**0.11**
pyridine N (%) [c]	**43.6**	**23.2**	**27.5**	**35.0**	**31.0**	**29.4**	**18.4**
quaternary N (%) [d]	**56.4**	**76.8**	**72.5**	**65.0**	**69.0**	**70.6**	**81.6**
$N_{pyridine}/N_{quaternary}$ [e]	**0.77**	**0.30**	**0.38**	**0.54**	**0.45**	**0.42**	**0.23**

[a] data from EA, [b, c, d] data from XPS, [e] data from c and d.

Table S2. Comparison of the LIBs performance of G-PC-800 and other reported cathode materials.

Sample	Condition	Voltage Window (V versus Li/Li$^+$)	Specific capacity (mA h g^{-1}) (Cycles)	Ref
PPy/r-GO	700 mA g^{-1}	2.0-4.0	61 (200)	1
PI-4	73.4 mA g^{-1}	1.5-3.5	173 (100)	2
LF-SWNT	200 mA g^{-1}	1.8-3.6	200 (100)	3
PPy/FC	50 mA g^{-1}	1.5-4.0	115 (100)	4
ACTF	5 A g^{-1}	1.5-4.5	80 (1000)	5
PI/SWNT	221.5 mA g^{-1}	1.5-3.5	175 (200)	6
BFFD	0.1 C	1.75-3.25	201 (200)	7
MWNT/graphene	100 mA g^{-1}	1.5-4.5	135 (100)	8
G-PC-800	**5 A g^{-1}**	**1.5-4.5**	**161 (3000)**	**This work**

Table S3. Comparison of the SIBs performance of G-PC-800 and other reported cathode materials.

Sample	Condition	Voltage Window (V versus Na/Na$^+$)	Specific capacity (mA h g^{-1}) (Cycles)	Ref
BPOE	1 A g^{-1}	1.3-4.1	90 (7000)	9
$Na_4C_8H_2O_6$	18.7 mA g^{-1}	1.6-2.8	183 (100)	10
PTCDA	200 mA g^{-1}	1.0-3.0	130 (195)	11
PTCDA-PI	0.1 C	1.5-3.5	148.9 (400)	12
$NaNiFe(CN)_6$	10 mA g^{-1}	2.5-3.8	65 (5)	13
$NaFePO_4$	0.05 C	1.5-4.5	142 (200)	14
R-FeHCF	300 mA g^{-1}	1.75-3.75	105 (1000)	15
NVP@C@rGO	100 C	2.5-3.8	55 (10000)	16
G-PC-800	**1 A g^{-1}**	**1.3-4.1**	**137 (500)**	**This work**

Table S4. Comparison of the capacitive performance of G-PC-800 and other reported electrode materials.

Sample	Condition	electrolyte	Specific gravimetric capacitances (F g^{-1})	Ref
RGO	0.1 A g^{-1}	6M KOH	41.5	17
Graphene (activated)	0.7 A g^{-1}	$BMIMBF_4$/AN	200	18
N-CNF-900	1 A g^{-1}	6M KOH	202	19
TNN-500	0.2 A g^{-1}	1 M H_2SO_4	285	20
ENM700	0.05 A g^{-1}	6M KOH	281	21
3D porous carbon	10 mV s^{-1}	1 M H_2SO_4	176	22
B/N-GAs	5.0 mV s^{-1}	1 M H_2SO_4	239	23
HGF-EC	1 A g^{-1}	$BMIMBF_4$/AN	298	24
G-PC-800	**0.1 A g^{-1}**	**6M KOH**	**340**	**This work**

Notes and references

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