

Supporting Information

Dual Atomic Defect Modulation in Three-Dimensional Mesoporous Graphene for High-Performance Potassium Ion Hybrid Capacitors

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Calculations:

Specific capacities (C_{cell-s}) of half-cells and PIHCs devices were estimated from the discharge curve using the following equations:

$$C_{cell-s} = \frac{\int_0^{\Delta t} I \times dt}{m}$$

(1)

where C_{cell-s} (mA h g⁻¹) is the specific capacity of the half-cells and PIHCs devices, I (mA) is the applied discharging current, Δt (h) is the discharging time and m (g) is the mass of the active material.

Specific energy density E and specific power density P of PIHCs devices were obtained from the following equations:

$$E = C_{cell-s} * \Delta V \quad (2)$$

$$P = \frac{C_{cell-s} * \Delta V}{\Delta t}$$

(3)

where E (Wh kg⁻¹) is the energy density, C_{cell-s} is the specific capacity obtained from Equation (1) and ΔV (V) is the voltage window. P (W kg⁻¹) is the specific power density and Δt (h) is the discharging time.

The chemical diffusion coefficient of k⁺ can be calculated based on the following equation:

$$D_{GITT} = \frac{4}{\pi\tau} \left(\frac{n_m V_m}{S} \right)^2 \left(\frac{\Delta E_s}{\Delta E_t} \right)^2 \quad (4)$$

Here, τ (s) is the duration of the current pulse; n_m (mol) is the number of moles; V_m (cm³ mol⁻¹) is the molar volume of the electrode; S (cm²) is the electrode/electrolyte contact area; ΔE_s is the steady state voltage change due to the current pulse, and ΔE_t is the voltage change during the constant current pulse, eliminating the iR drop.



Figure S1. NP-3DPG powder on an evaporating dish.

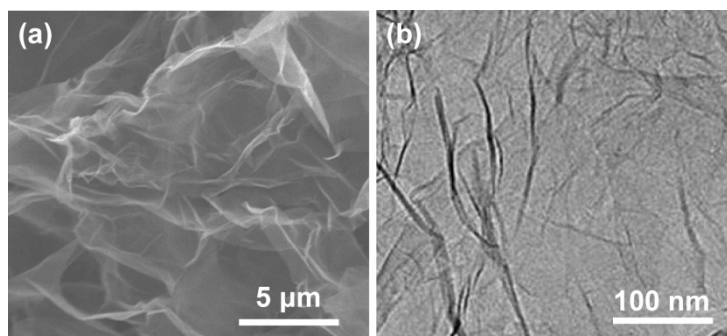


Figure S2. (a) SEM and (b) TEM of GO.

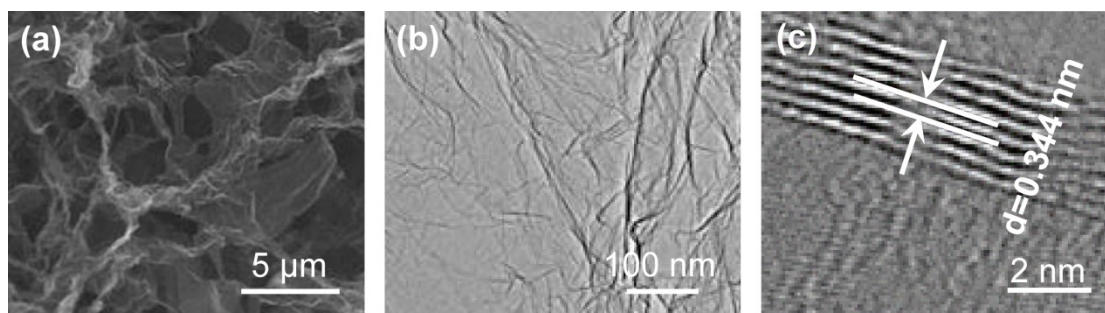


Figure S3. (a) SEM, (b) TEM, and (C) HRTEM of N-3DPG.

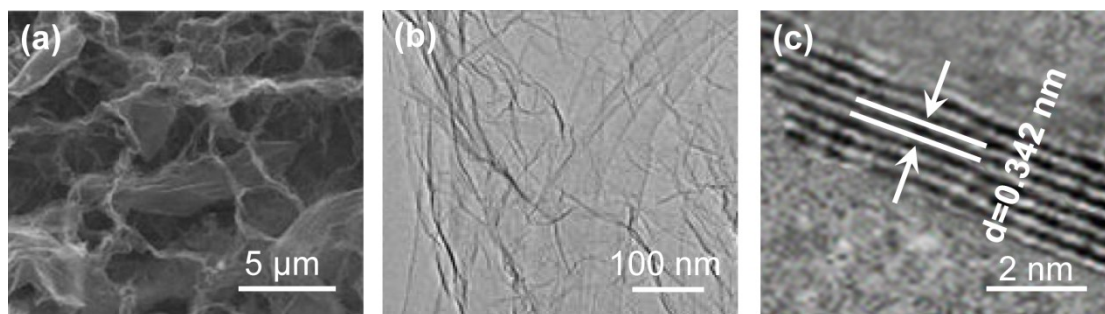


Figure S4. (a) SEM, (b) TEM, and (c) HRTEM of 3DPG.

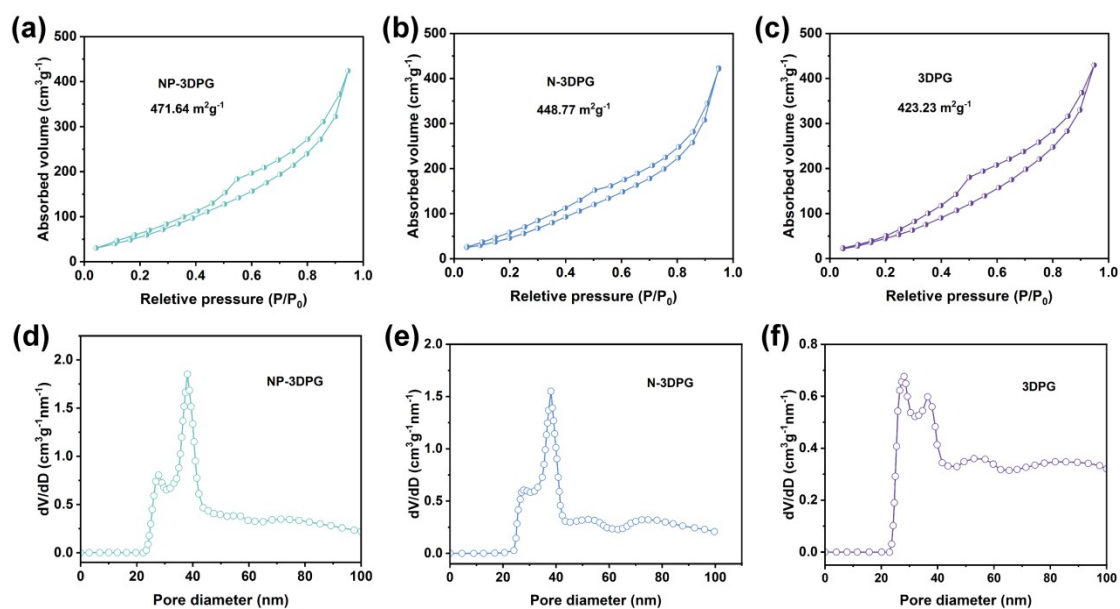


Figure S5. Nitrogen adsorption–desorption isotherm of (a) NP-3DPG, (b) N-3DPG, and (c) 3DPG. Pore size distribution of (d) NP-3DPG, (e) N-3DPG, and (f) 3DPG.

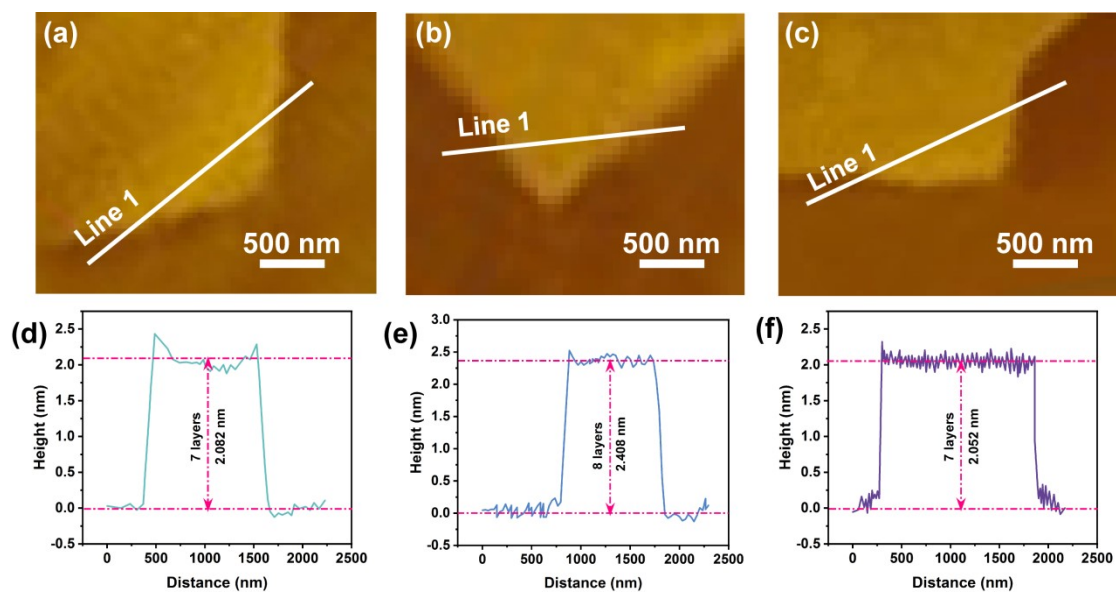


Figure S6. AFM images of (a) NP-3DPG, (a) N-3DPG, and (c) 3DPG. Height profiles of (d) NP-3DPG, (e) N-3DPG, and (f) 3DPG.

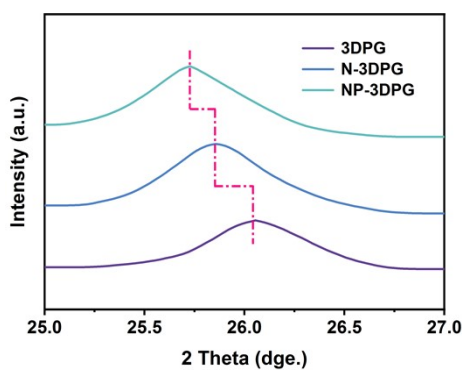


Figure S7. Partial enlarged XRD patterns of 3DPG, N-3DPG and NP-3DPG.

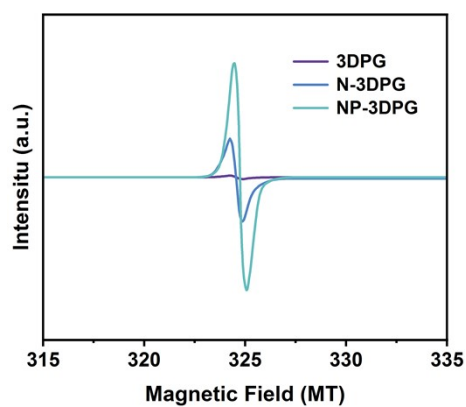


Figure S8. EPR spectra of NP-3DPG, N-3DPG and 3DPG.

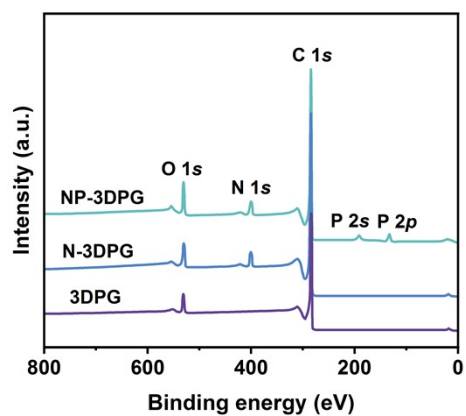


Figure S9. XPS survey spectrum of 3DPG, N-3DPG, and NP-3DPG.

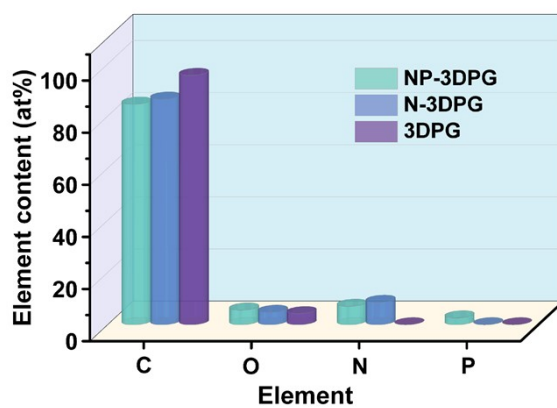


Figure S10. Element content of 3DPG, N-3DPG, and NP-3DPG.

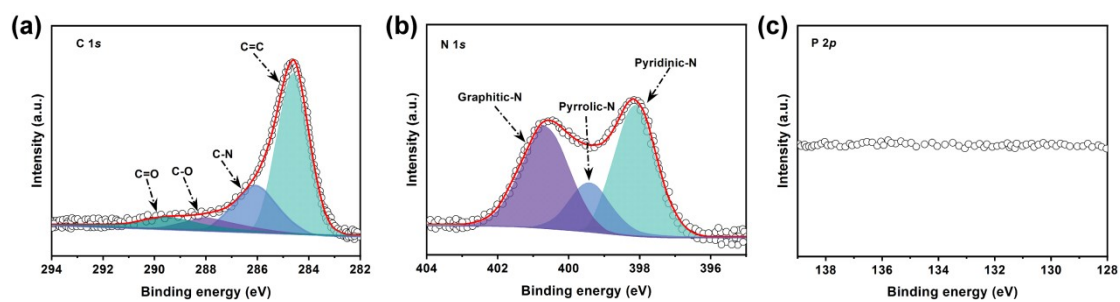


Figure S11. (a) C 1s, (b) N 1s, and (c) P 2p high-resolution XPS of N-3DPG.

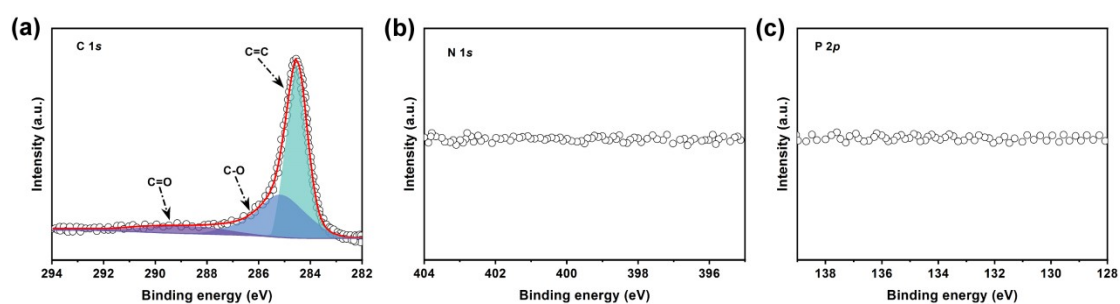


Figure S12. (a) C 1s, (b) N 1s, and (c) P 2p high-resolution XPS of 3DPG.

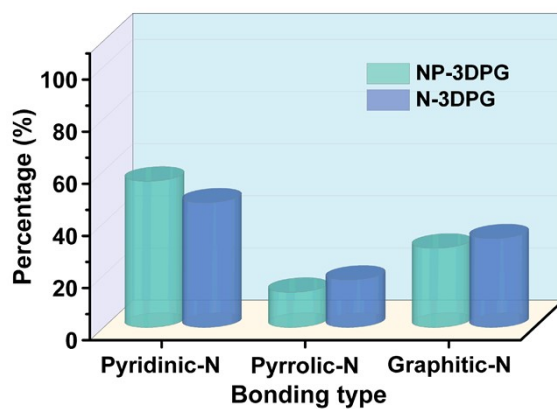


Figure S13. Percentage contents of nitrogen species in the N-3DPG and NP-3DPG.

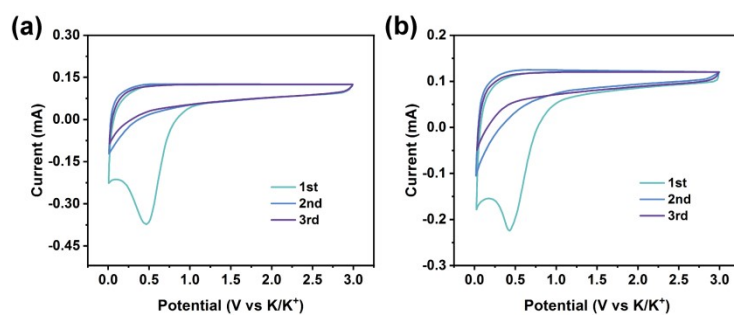


Figure S14. CV curves of (a) N-3DPG and (b) 3DPG electrode.

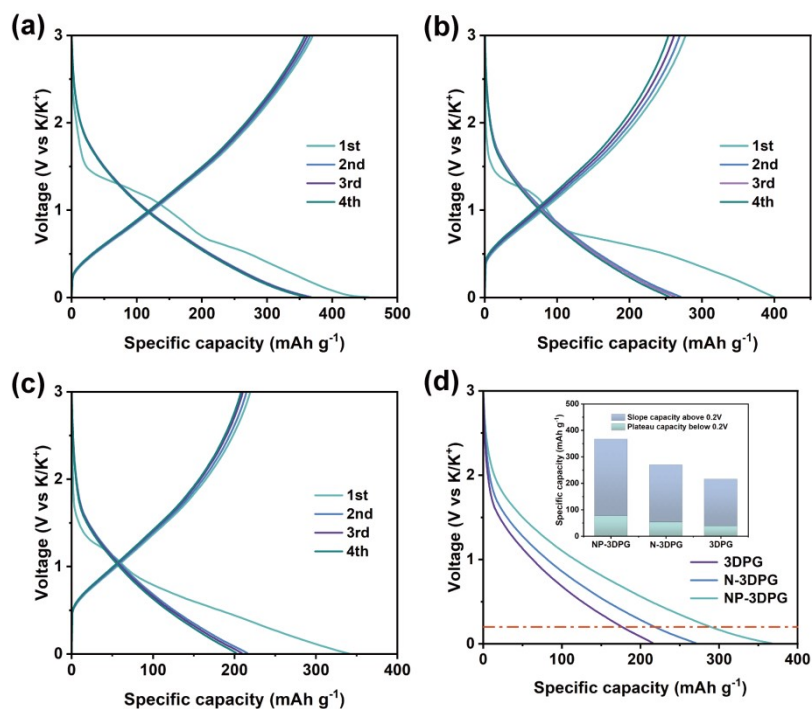


Figure S15. GCD profiles of the (a) NP-3DPG, (b) N-3DPG and (c) 3DPG electrodes at 0.2 A g^{-1} . (d) The discharge profiles of NP-3DPG, N-3DPG and 3DPG at second cycle (inset: the capacity contribution at each stage).

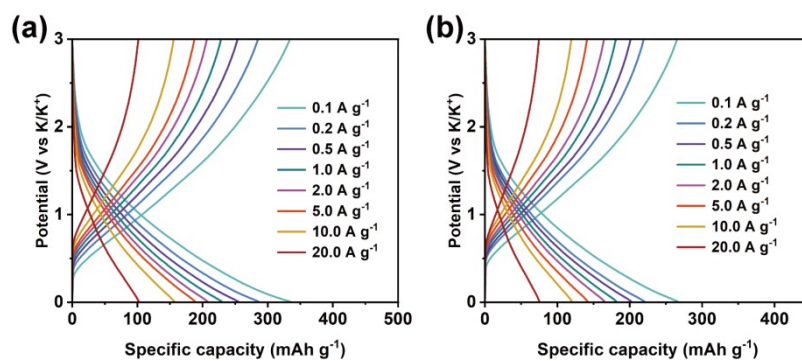


Figure S16. GCD curves at various current densities for (a) N-3DPG and (b) 3DPG.

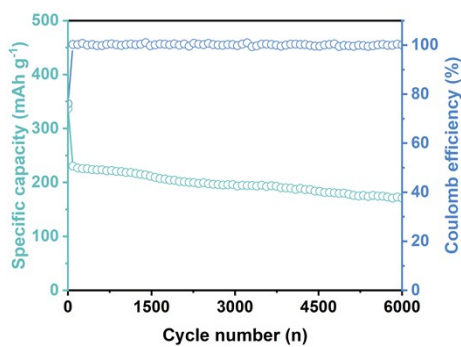


Figure S17. Long-term cycling performance of the NP-3DG at 5.0 A g⁻¹.

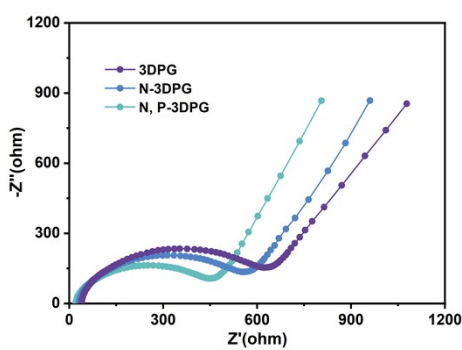


Figure S18. Electrochemical impedance spectra (EIS) of the 3DPG, N-3DPG and NP-3DPG electrodes.

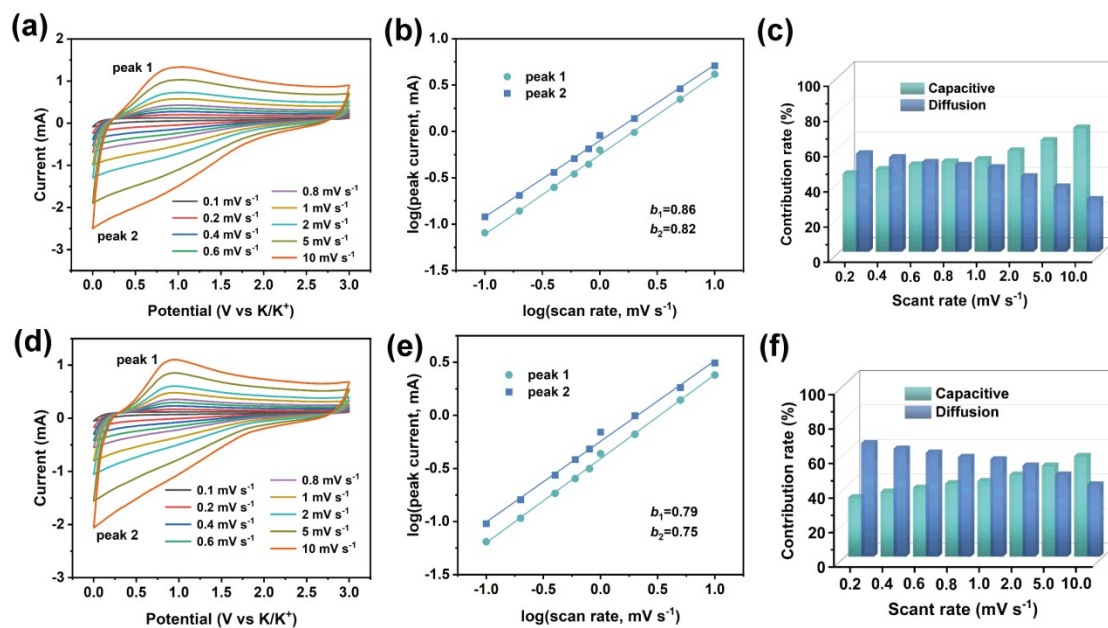


Figure S19. CV curves of (a) N-3DPG and (d) 3DPG. b -value of (b) N-3DPG and (e) 3DPG. Diffusive and capacitive contributions of (c) N-3DPG and (f) 3DPG.

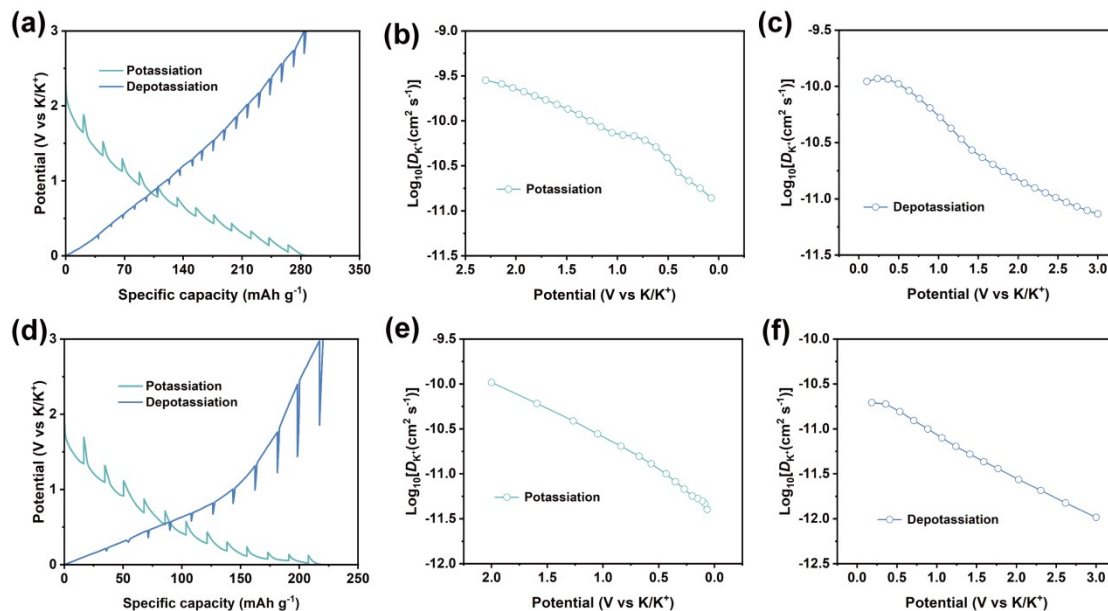


Figure S20. GITT profiles of (a) N-3DPG and (d) 3DPG; Potassiation D_k of (b) N-3DPG and (e) 3DPG; and depotassiation D_k of (c) N-3DPG and (f) 3DPG.

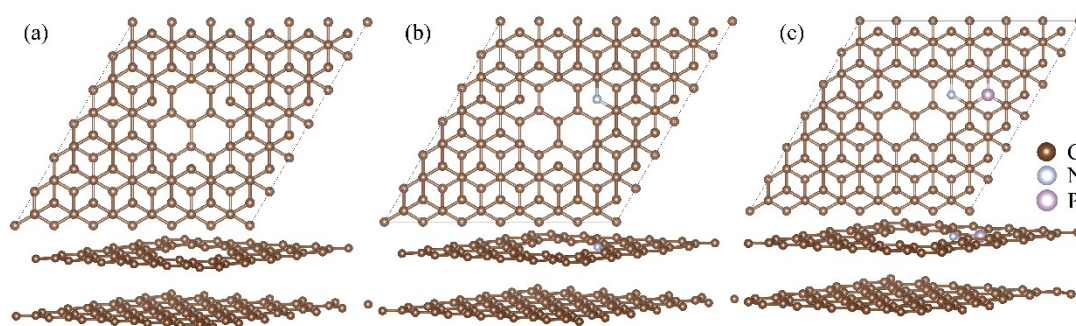


Figure S21. Top and side view of the optimized structure of (a) 3DPG, (b) N-3DPG and (c) NP-3DPG.

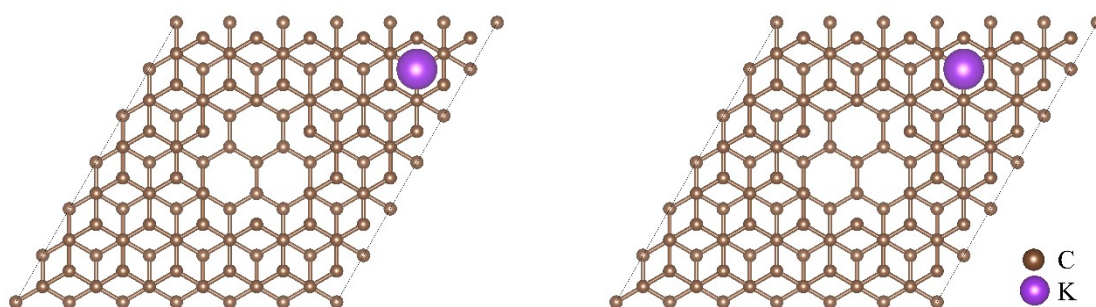


Figure S22. Illustration of the K ion diffusion path from one hollow position to the nearest hollow position in 3DPG.

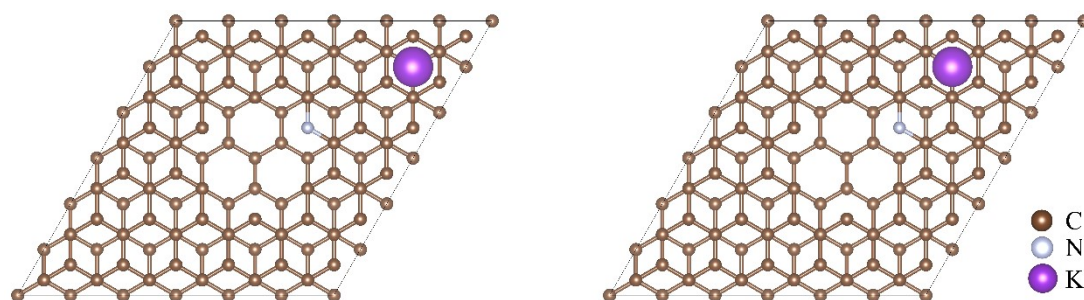


Figure S23. Illustration of the K ion diffusion path from one hollow position to the nearest hollow position in N-3DPG.

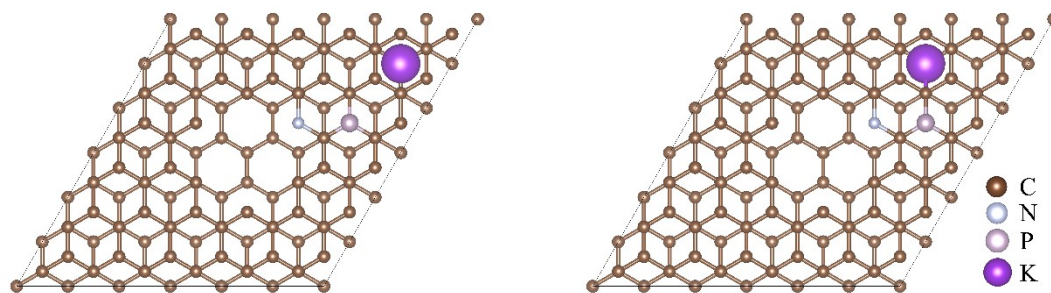


Figure S24. Illustration of the K ion diffusion path from one hollow position to the nearest hollow position in NP-3DPG.

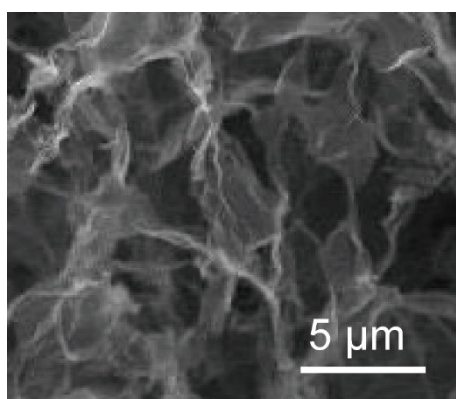


Figure S25. SEM of ANP-3DPG.

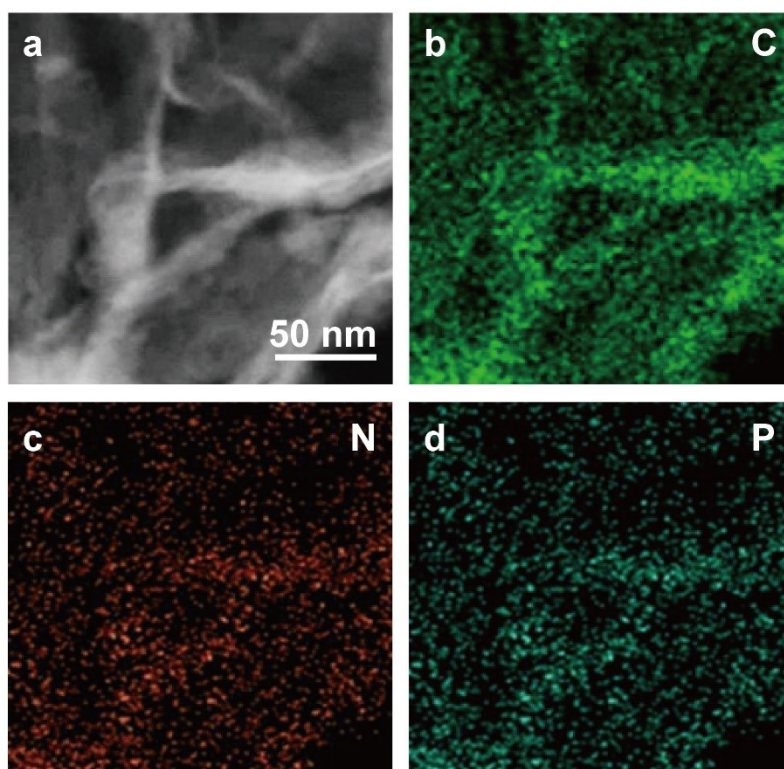


Figure S26. (a) STEM image, (b) C, (c) N, and (d) P elemental mapping of ANP-3DPG.

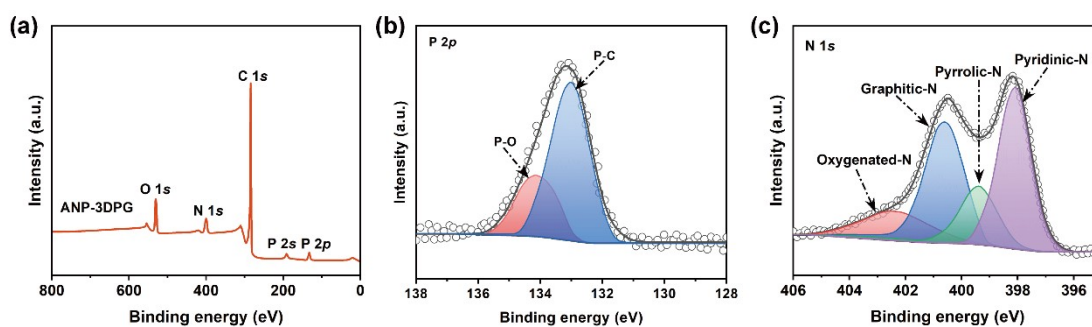


Figure S27. (a) The survey XPS spectrum, (b) high-resolution P 2p spectra, (c) high-resolution N 1s spectra of ANP-3DPG.

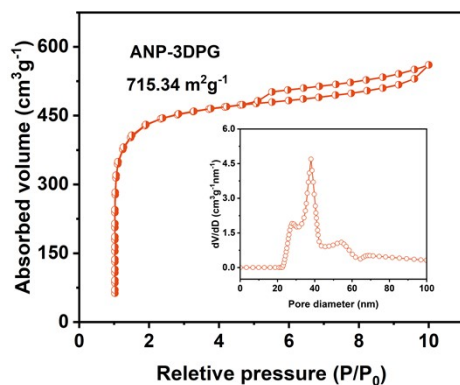


Figure S28. Nitrogen adsorption–desorption isotherm of ANP-3DPG.

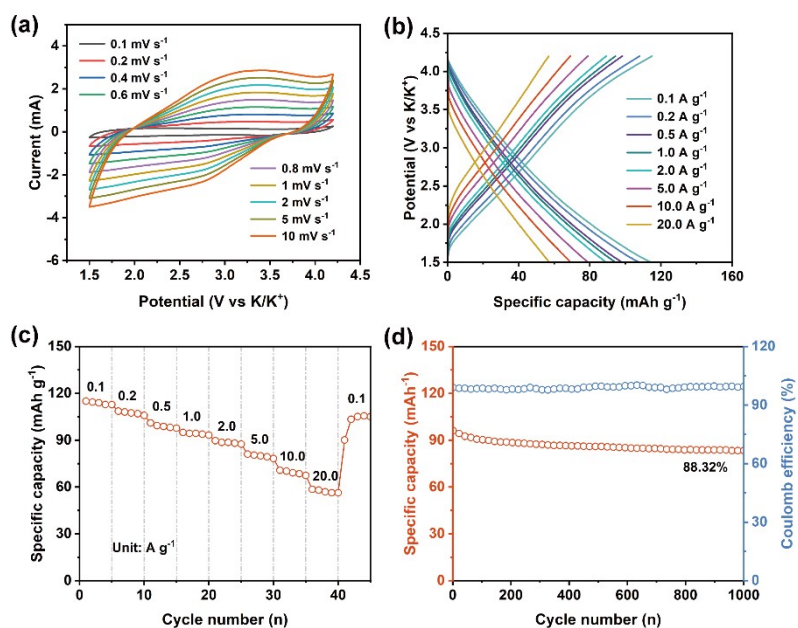


Figure S29. Electrochemical performances of the ANP-3DPG cathode in K-half cells.

(a) CV curves at different scan rates, (b) charge/discharge curves at various current densities, (c) rate property, and (g) cycling performance at 1.0 A g⁻¹.

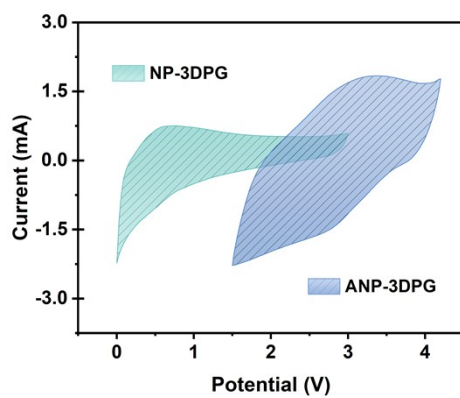


Figure S30. CV curves of the NP-3DPG and the ANP-3DPG electrodes in half cells at 1 mV s^{-1} .

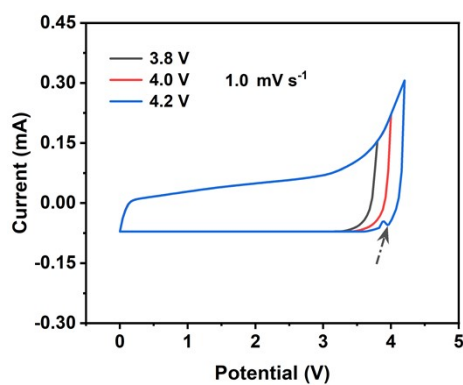


Figure S31. CV curves of ANP-3DPG//NP-3DPG PIHCs device in different potential windows at a scan rate of 1.0 mV s^{-1} .

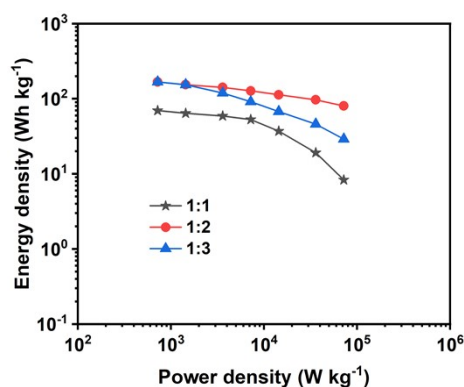


Figure S32. Ragone plots of NP-3DPG//ANP-3DPG PIHCs device with different mass ratios of anode to cathode.

Table S1. Comparison of the initial coulombic efficiency of the carbonaceous electrodes for PIBs with our work.

Materials	Current density (A g ⁻¹)	Initial coulombic efficiency	Reference
NP-3DPG	0.2	81.4%	This work
PN-HPCNF	0.2	78.9%	[1]
PDDA-NPCNs/Ti ₃ C ₂	0.1	73.2%	[2]
nitrogen/oxygen codoped carbon hollow multihole bowls	0.1	63.3%	[3]
oxygen/fluorine dualdoped porous carbon	0.1	63%	[4]

S/O codoped porous carbon microspheres	0.05	61.7%	[5]
mesoporous graphitic carbon nanospring	0.05	61.2%	[6]
P-doped N-rich honeycomb-like carbon	0.2	56.9%	[7]
Sulfur-grafted hollow carbon spheres	0.025	51.4%	[8]
N-doped carbon nanofibers-650	0.025	49%	[9]
N/O dual-doped carbon network	0.05	47.12%	[10]
P-doped hard carbon	0.1	45.7%	[11]
rGO aerogel	0.1	44%	[12]
nitrogen-doped porous carbon	0.1	43.1%	[13]
graphitic carbon nanocage	0.05	40%	[14]
carbon nanosheets750	0.05	38%	[15]
nitrogen/oxygen in situ dual-doped hierarchical porous hard carbon	0.05	25%	[16]
S/N@C	0.05	24.6%	[17]

three-dimensional nitrogen-doped framework	0.1	24.3%	[18]
carbon NHC ₂ -NH ₃ /Ar	0.1	15.8%	[19]
Hierarchical carbon nanotube-S4	0.1	15%	[20]
pyridinic N-contentdoped porous carbon monolith	0.02	10%	[21]

Table S2. Potassium storage performance of NP-3DPG compared with previously reported materials.

Materials	Rate capacity	Cycling stability	Ref.
NP-3DPG	429.5 mA h g ⁻¹ at 20.0 A g ⁻¹	236.5 mA h g ⁻¹ after 15000 cycles at 5.0 A g ⁻¹	This work
Nitrogen doped cup-stacked carbon tubes	75 mA h g ⁻¹ at 1 A g ⁻¹	236 mA h g ⁻¹ after 100 cycles at 0.02 A g ⁻¹	[22]
High pyridine N- doped porous carbon	182.6 mAh g ⁻¹ at 2 A g ⁻¹	231.6 mAh g ⁻¹ after 200 cycles at 0.5 A g ⁻¹	[23]
Co ₃ O ₄ -Fe ₂ O ₃ /C	278 mAh g ⁻¹ at 1 A	220 mAh g ⁻¹ after 50 cycles	[24]

	g^{-1}	at 0.05 A g^{-1}	
Hyperporous carbon sponge	180 mAh g^{-1} at 1.6 A g^{-1}	210 mAh g^{-1} after 500 cycles at 1 A g^{-1}	[20]
Nitrogen-doped bamboo-like carbon nanotubes	186 mAh g^{-1} at 1 A g^{-1}	204 mAh g^{-1} after 1000 cycles at 0.5 A g^{-1}	[25]
Chemically activated hollow carbon nanospheres	137 mA h g^{-1} at 4 A g^{-1}	192.7 mAh g^{-1} after 5000 cycles at 2 A g^{-1}	[26]
Sulfur/nitrogen codoped carbon nanofiber agerogel	168 mA h g^{-1} at 2 A g^{-1}	168 mA h g^{-1} after 1000 cycles at 2 A g^{-1}	[27]
Hollow carbon architecture	110 mAh g^{-1} at 0.56 A g^{-1}	150 mA h g^{-1} after 500 cycles at 0.279 A g^{-1}	[28]
Nitrogen doped carbon nanofibers	153 mA h g^{-1} at 2 A g^{-1}	146 mA h g^{-1} after 4000 cycles at 2 A g^{-1}	[9]
S-NC	72 mA h g^{-1} at 10 A g^{-1}	140 mA h g^{-1} after 3000 cycles at 2 A g^{-1}	[29]
3D nitrogen-doped framework carbon	111 mA h g^{-1} at 10 A g^{-1}	137 mA h g^{-1} after 1000 cycles at 2 A g^{-1}	[18]
N-doped carbon	$102.6 \text{ mA h g}^{-1}$ at 2 A g^{-1}	$119.9 \text{ mA h g}^{-1}$ after 1000 cycles at 1 A g^{-1}	[30]

Oxygen-Rich	133 mA h g ⁻¹ at 10	111 mA h g ⁻¹ after 3000	[15]
Carbon Nanosheets	A g ⁻¹	cycles at 5 A g ⁻¹	
Onion-like carbon	78 mA h g ⁻¹ at 10	111 mA h g ⁻¹ after 1000	[31]
	A g ⁻¹	cycles at 2 A g ⁻¹	
Sulfur/oxygen	158 mAh g ⁻¹ at 1 A	108.4 mA h g ⁻¹ after 2000	[5]
co-doped hard	g ⁻¹	cycles at 1 A g ⁻¹	
carbon			
Nitrogen-doped	102 mA h g ⁻¹ at 2	102 mA h g ⁻¹ after 500	[32]
carbon nanotubes	A g ⁻¹	cycles at 2 A g ⁻¹	
Hierarchically	64 mA h g ⁻¹ at 4 A	65 mA h g ⁻¹ after 900	[17]
Porous Thin	g ⁻¹	cycles at 2 A g ⁻¹	
Carbon Shells			

Table S3. The comparison between NP-3DPG//ANP-3DPG PIHCs and other recently reported carbonaceous based PIHCs.

Devices	Maximum	Maximum	Cycles	Ref.
	Energy density (Wh kg ⁻¹)/Power density (W kg ⁻¹)	Power density (W kg ⁻¹)/Energy density (Wh kg ⁻¹)	/Capacity retention (%)/current (A g ⁻¹)	
MoP@NC-1//AC	69.7/100	2041.6/20	800/89.9/0.1	[33]

CTP@C//AC	80/32	5144/34	1000/75.9/5	[34]
NOHPC//HPAC	90.1/939.6	6217.5/52.93	6000/87.2/2	[35]
BSH//AC	94/188	599/13.3	1000/71.4/0.34	[36]
PNC//HPC	103/97.9	6106.2/44.1	5000/64/1	[37]
WS ₂ @NCNs//NCHS	103.4/235	2300/60	500/78/0.5	[38]
NHCS//ANHCS	114.2/100.5	8203/19.1	5000/80.4/2	[39]
NCNT//AC	115/116.8	1713.4/25.7	2000/81.6/1	[40]
N-MoSe ₂ /G//AC	119/39.6	7212/29	3000/75.2/1	[41]
N,S-3DHPC-600//AC-800	130.6/210	16800/56	5000/86.8/5	[42]
NbSe ₂ /NSeCNFs//AC	133/180	4000/18	10000/83/2	[43]
SHPNC//AC	135/112.6	~4000/20	3750/75.4/1	[44]
PN-PanC//PN-PanC	155.9/76.1	11309.1/22.0	40000/77/1	[45]
M-NC/ANC	157/37	2300/49	3000/80//0.5	[46]
P/O-PCS//AC	158/223	1380/11	30000/94.5/5	[47]
3DNFC//3DNFAC	163.5/210	21000/76.4	10000/91.7/2	[18]
NP-3DPG//ANP-3DPG	167.4/720	72000/80.2	10000/87.3/2	This work

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