Phytic Acid Induced Nitrogen Configuration Adjustment of Active Nitrogen-Rich Carbon Nanosheets for High-Performance Potassium-Ion Storage

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1. Assembly of full battery

The FeHCF, Super P and polyvinylidene fluoride were mixed with a ratio of 8:1:1 in water to form a homogeneous slurry. After that, the homogeneous slurry was coated on Al-C foil and then dry in vacuum oven over night. The resulting electrode was cut into discs with a diameter of 14 mm as the cathode. Before the assembly of the full K-ion battery, the anode and the cathode were first run 2 cycles to guarantee the pretreatment of both electrodes. After that, the capacity of the cathode and the anode were matched for the assembly of the full K-ion battery with the same electrolyte in the half cell under Ar atmosphere.

2 DFT computational methods

Theoretical calculations were performed on the basis of density functional theory (DFT), as implemented in the Vienna ab-initio simulation package (VASP). The Perdew-Becke-Ernzerhof (PBE) functional and projector augmented wave (PAW) scheme were adopted for geometric optimizations. All structures were optimized with a convergence criterion of 1×10^{-4} eV for the energy and 0.05 eV/Å for the forces. The plane wave expansion was chosen and cut-off at the kinetic energy of 500 eV. In addition, the Brillouin zones were sampled by a ($3 \times 3 \times 3$) grid generated with Monkhorst-Pack scheme. For the adsorption energy calculation according to the calculation formula: $\Delta E_a = E_{total} - E_{mater} - E_K$, where E_{total} was the total energy of the K-ion adsorbed compounds, E_{mater} was the energy of nitrogen-doping carbon, E_K was the energy of the K-ion. The K-ion diffusion was calculated by the NEB method. The minimum energy path (MEP) for K diffusion from one interstitial site to its

neighboring one was searched by linear synchronous transit (LST)/quadratic synchronous transit (QST) method. The diffusion barrier was defined as the energy difference between the peak image and the initial state.

3 Diffusion coefficient calculated methods

The K-ion diffusion coefficients in solid-state bulks were calculated according to the data in the low frequency as follows:

$$D = \frac{R^2 T^2}{2n^2 A^2 F^4 C^2 \delta^2}$$
(1)

Here, R is the gas constant with a value of 8.314 J mol⁻¹ K⁻¹, T means the test temperature (298 K), A is the area of electrode (1.54 cm²), n represents the transferred electron number with a value of 1, F stands for the Faraday constant (96485 C mol⁻¹) and C means the volume concentration (~0.8 mmol cm⁻³). δ can be calculated from the slope of the plot of Z' against $\omega^{-1/2}$ in the low-frequency region.

Table S1	The obtained	parameters of	adsor	ption mod	els	based	l on I	DFT	calculation.
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System	E _{total} (eV)	E _{mater} (eV)	E _K (eV)	ΔE_a (eV)	
graphitic-N	-462.13	-461.12	-0.038	-0.98	
pyrrolic-N	-404.59	-400.53	-0.038	-4.02	
pyridinic-N	-394.95	-390.27	-0.038	-4.64	
oxidized-N	-400.41	-396.13	-0.038	-4.24	
2-pyridinic-N	-396.43	-391.67	-0.038	-4.72	

System		Site 1	Site 2	Site 3	Site 4	Site 5	Site 6	Site 7
pyrrolic-N	E _{total} (eV)	-404.594	-404.470	-403.875	-403.476	-403.866	-404.471	-404.599
	E _{bar} (eV)	0	0.124	0.719	1.118	0.728	0.123	-0.005
pyridinic-N	E _{total} (eV)	-394.951	-394.915	-394.833	-394.792	-394.836	-394.917	-394.951
	E _{bar} (eV)	0	0.036	0.118	0.159	0.115	0.034	0

Table S2 The obtained parameters of NEB models based on DFT calculation.



Figure S1 Electronic localization function of a) graphitic-N, (b) pyrrolic-N, (c)

pyridinic-N, (d) oxidized-N. The brown, blue grey, red and blue-violet balls represent

the C, N, O, and K atoms, respectively.



Figure S2 SEM images of (a) PANC-1 and (b) PANC-3



Figure S3 The SEM images of (a) NC, (b) RGO and (c) PN. Obviously, without

graphene as a template, the morphology of the material becomes massive.



Figure S4 The XRD patterns of NC, RGO and PN. The results show that they are also

amorphous carbon.



Figure S5 XRD of PANCs.



Figure S6 Raman of PANCs.



Figure S7 (a) Nitrogen adsorption–desorption isotherm and (b) mesoporous size distribution curves of PANCs. The specific surface areas of PANC-1, PANC-2 and

PANC-3 are 19.97, 21.52 and 25.31 $m^2 \ g^{\text{-1}}$, respectively.



Figure S8 Survey XPS spectrum of PANCs.



Figure S9 Cycling performance of PANC-2-300 (annealing at 300 °C), PANC-2 and

PANC-2-700 (annealing at 700 °C).



Figure S10 The cycle performances of NC, RGO and PN. Obviously, the potassium storage performances of these materials are significantly worse than the optimized

PANCs.



Figure S11 Partial enlargement EIS in (a) high frequency region and (b) intermediate

frequency region.



Figure S12 CV curves and at different scan rates from 0.2 to 1.0 mV s⁻¹ of PANC-1,

PANC-2 and PANC-3.



Figure S13 The lighted blue LED panel driven by the full battery.