

## Supporting Information

# Two-Dimensional Molecular Brush-Based Ultrahigh Edge-Nitrogen-Doped Carbon Nanosheets for Ultrafast Potassium-Ion Storage

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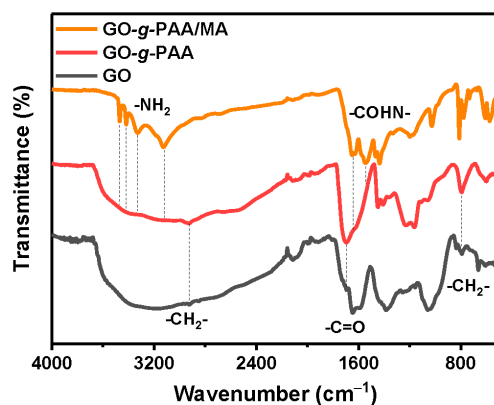


Figure S1. FT-IR spectra of GO, GO-g-PAA and GO-g-PAA/MA.

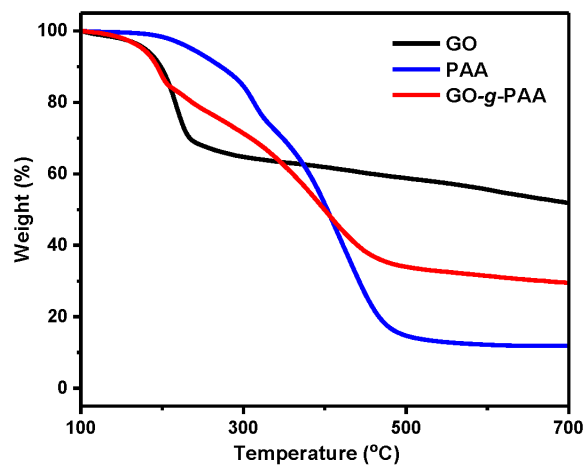
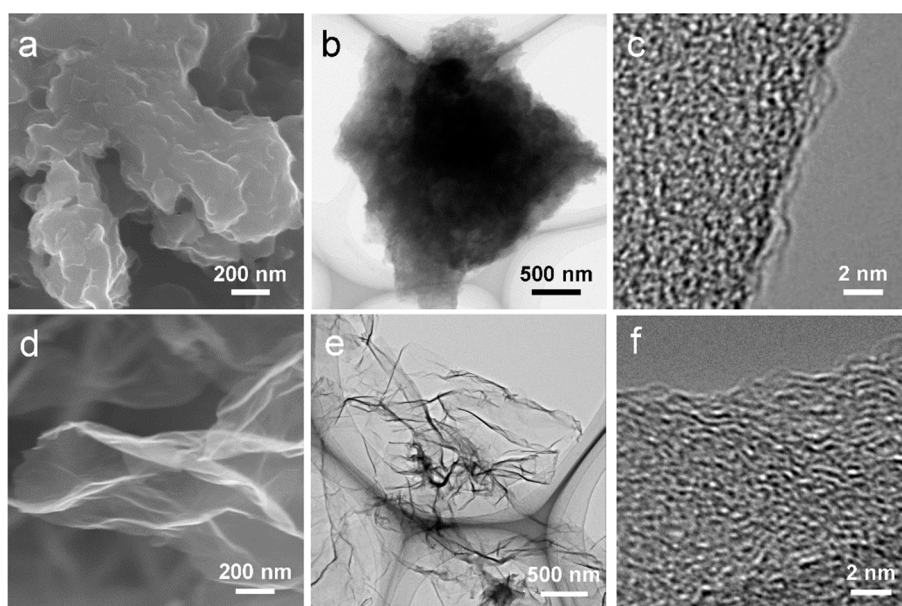
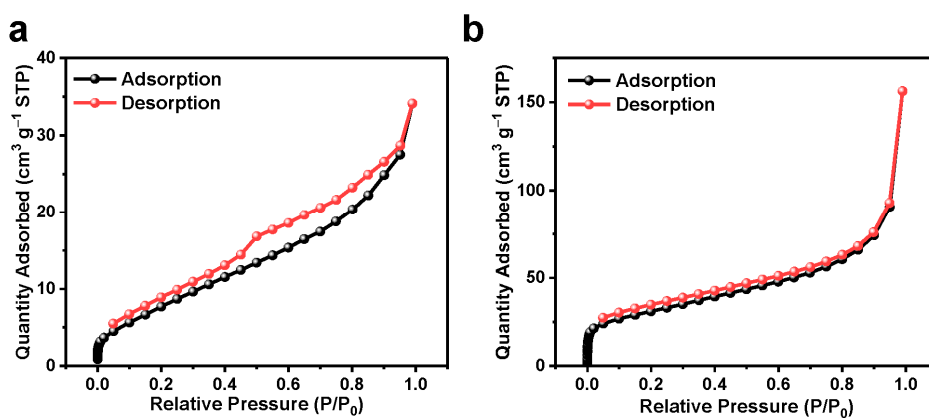


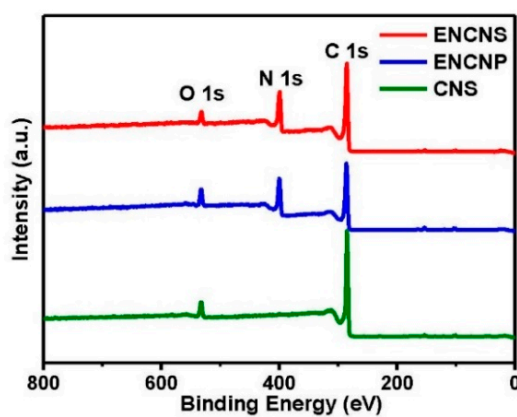
Figure S2. TGA curves of GO, PAA and GO-g-PAA.



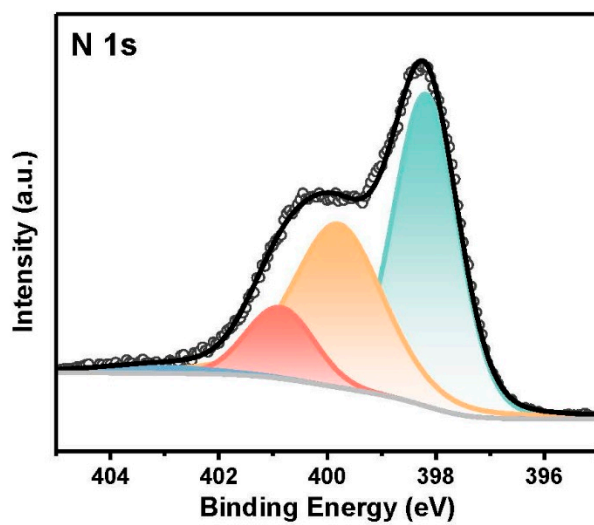
**Figure S3.** SEM, TEM and HRTEM images of (a-c) ENCNP and (d-f) CNS.



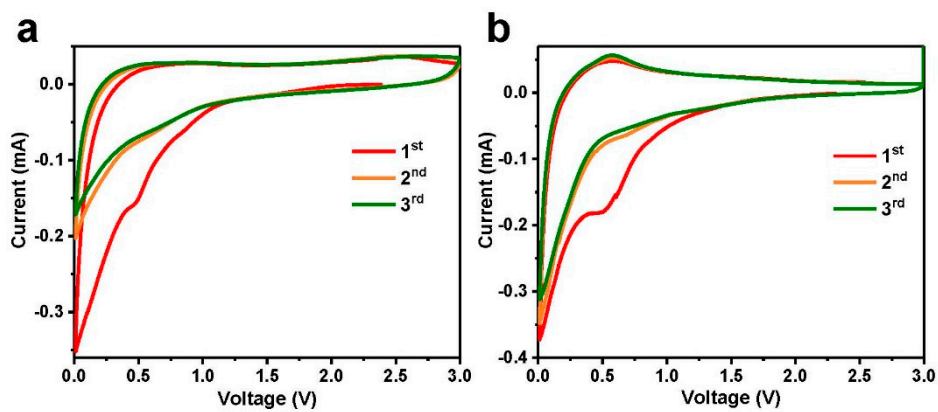
**Figure S4.** N<sub>2</sub> adsorption-desorption isotherms of (a) ENCNP and (b) CNS.



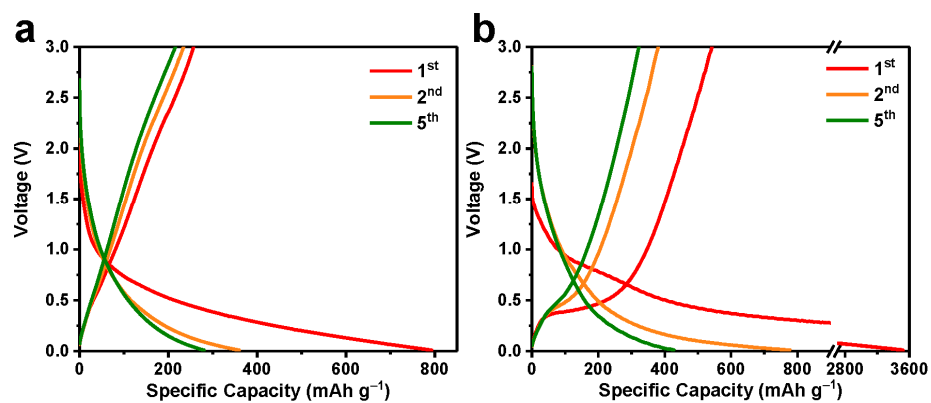
**Figure S5.** XPS full scans of ENCNS, ENCNP and CNS, revealing the presence of C, N and O elements.



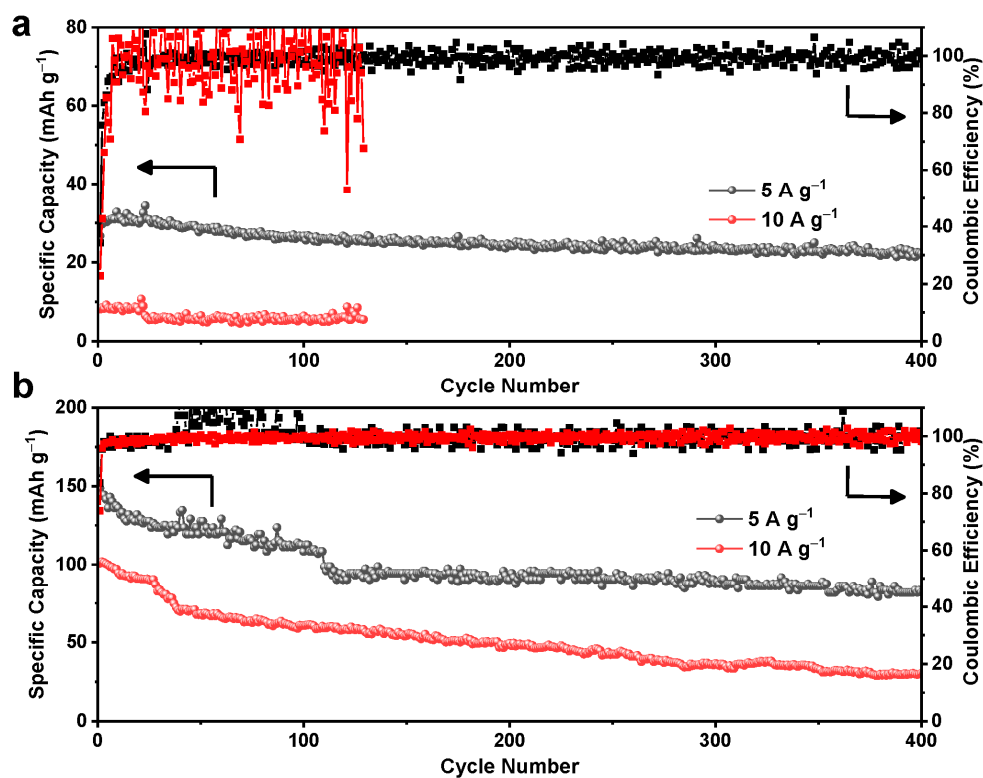
**Figure S6.** High-resolution N 1s XPS spectrum of ENCNP.



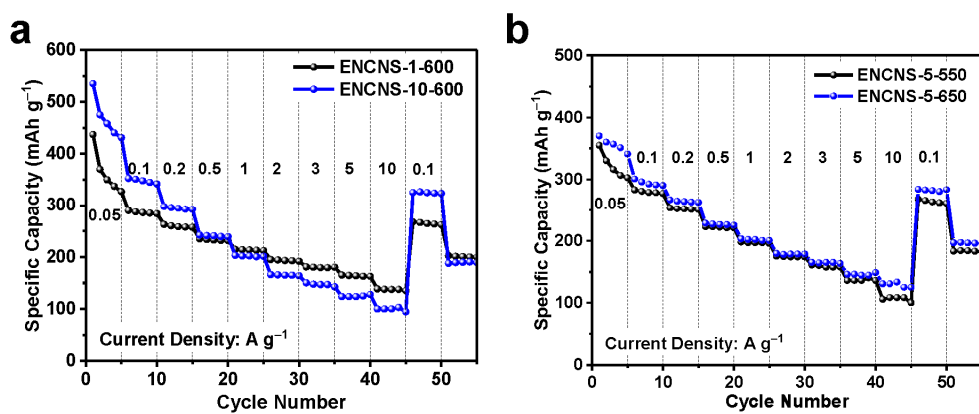
**Figure S7.** CV curves of (a) ENCNP and (b) CNS anodes at a scan rate of  $0.2 \text{ mV s}^{-1}$ .



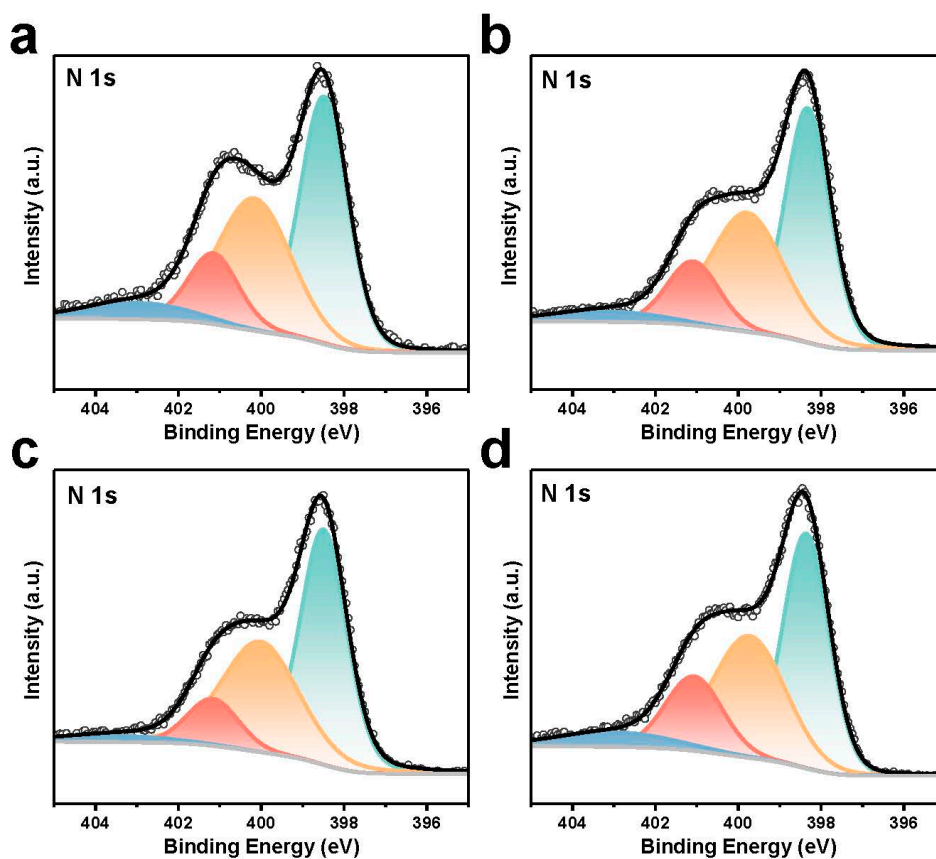
**Figure S8.** Discharge/charge curves of (a) ENCNP and (b) CNS anodes at a current density of  $0.05 \text{ A g}^{-1}$ .



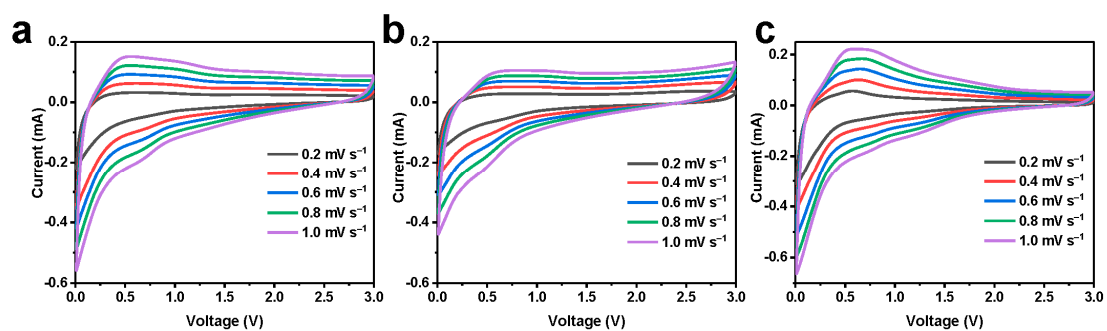
**Figure S9.** Long-term cycling performances of (a) ENCNP and (b) CNS at 5 and 10 A g<sup>-1</sup>.



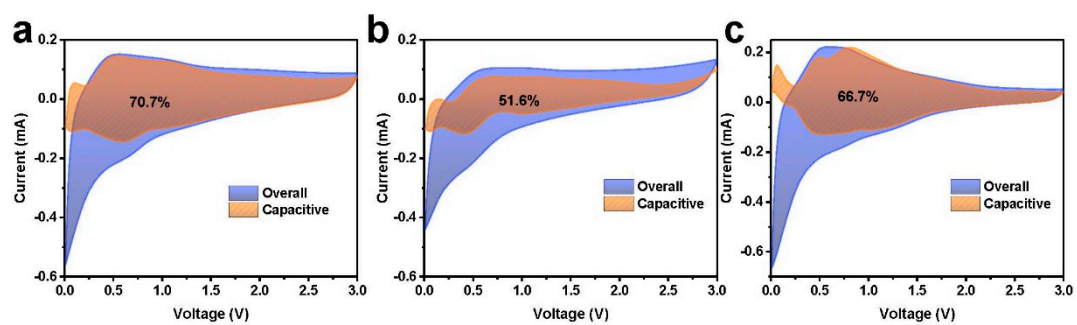
**Figure S10.** Rate capability of (a) ENCNS-1-600 and ENCNS-10-600 as well as (b) ENCNS-5-550 and ENCNS-5-650 at various current densities between 0.05 and 10.0 A g<sup>-1</sup>.



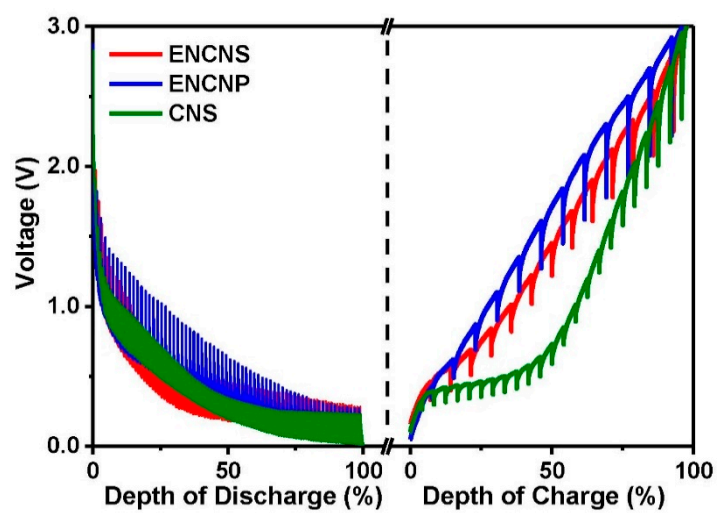
**Figure S11.** High-resolution N 1s XPS spectra of (a) ENCNS-1-600, (b) ENCNS-10-600, (c) ENCNS-5-550, and (d) ENCNS-5-650.



**Figure S12.** CV curves of (a) ENCNS, (b) ENCNP, and (c) CNS at scan rates of 0.2, 0.4, 0.6, 0.8 and 1.0  $\text{mV s}^{-1}$ .



**Figure S13.** Capacitive contributions of (a) ENCNS, (b) ENCNP, and (c) CNS at a scan rate of 1.0 mV s<sup>-1</sup>.



**Figure S14.** GITT potential profiles of ENCNS, ENCNP, and CNS.

**Table S1.** Total nitrogen and edge-nitrogen contents of various samples obtained with different molar ratios of -NH<sub>2</sub>/-COOH and heat treatment temperatures.

<b>Sample</b>	<b>Total nitrogen content (at%)</b>	<b>N6 content (at%)</b>	<b>N5 content (at%)</b>	<b>NQ content (at%)</b>	<b>NO content (at%)</b>	<b>Edge- nitrogen content (at%)</b>
<b>ENCNS</b>	20.6	8.5	7.7	2.8	1.6	16.2
<b>CNS</b>	0.6	-	-	-	-	-
<b>ENCNP</b>	22.4	10.9	8.7	2.4	0.4	19.6
<b>ENCNS-5-550</b>	24.1	11.6	9.3	2.5	0.7	20.9
<b>ENCNS-5-650</b>	19.3	8.1	6.7	3.2	1.3	14.8
<b>ENCNS-1-600</b>	9.3	4.1	3.4	1.3	0.5	7.5
<b>ENCNS-10-600</b>	21.6	9.4	8.3	3.0	0.9	17.7

**Table S2.** Total nitrogen content, edge-nitrogen content, and reversible capacity at 5 A g<sup>-1</sup> of ENCNS and other reported nitrogen-doped carbonaceous anode materials in PIBs.

Sample	Total nitrogen content (at%)	Edge-nitrogen content (at%)	Reversible capacity at 5 A g <sup>-1</sup> (mAh g <sup>-1</sup> )	Reference
ENCNS	20.6	16.2	168	<b>This work</b>
HNC	17.03	15.70	93.0	[1]
N-CNFs	13.1	7.2	144	[2]
NPPC-2-700	3.68	2.82	133	[3]
MCS-7-900	5.27	1.96	107.9	[4]
NCSN	8.10	5.91	149	[5]
ENDC	10.5	9.2	74	[6]
NSC	2.09	1.57	115	[7]
HENC	5.78	4.38	101	[8]
NCTP	11.7	9.0	134.7	[9]
HOCNs	16.54	12.58	136	[10]
N-PCNS	12.1	11.8	127.5	[11]
3DNFC	5.74	5.02	115	[12]
NCNFs	13.8	7.7	126	[13]



## Reference

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