

## Supporting Information

### **Phosphorus Doping Induced Kinetics Modulation for Nitrogen Doped Carbon Mesoporous Nanotubes as Superior Alkali Metal Anode Beyond Lithium for High Energy Potassium-Ion Hybrid Capacitors**

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## Experimental Section

**Synthesis of PNC-MeNTs:** In a typical process, 3 mmol of  $\text{MnSO}_4 \cdot \text{H}_2\text{O}$ , 2 mmol of  $\text{KMnO}_4$  and 2 mmol  $\text{NH}_4\text{F}$  were orderly added into 40 mL of deionized water, respectively. The solution was transferred to a 50 mL Teflon-lined autoclave and kept at 150 °C for 12 h, then allowed to naturally cooled to room temperature. The product of  $\text{MnO}_2$  was collected by centrifugation and washed with deionized water and ethanol. Subsequently, the pyrrole (0.38 mL) and aniline (0.29 mL) monomers were dissolved in 50 mL of  $\text{H}_2\text{SO}_4$  solution (0.1 M) and phytic acid solution (PA, 0.5 M), in which the molar ratio of PA to pyrrole and aniline is 1:1. The  $\text{MnO}_2$  nanowires (100 mg) were dispersed into deionized water by sonication 10 minutes and then added to the pyrrole/aniline/phytic acid (Py/AN/PA) solution under stirring for 4 h at room temperature. The obtained precipitate (denoted as PPy-PANI-PA) was washed with deionized water and ethanol several times and dried at 80 °C overnight. The black precursor was calcinated in a tubular furnace at 700 °C for 2 h with a ramp of 2 °C min<sup>-1</sup> in Ar atmosphere to form the final product of P/N codoped carbon mesoporous nanotubes (denoted as PNC-MeNTs). Meanwhile, the precursor PPy-PANI-PA also calcinated at other temperatures (650 and 800 °C) under Ar atmosphere. Besides, the nitrogen-doped carbonaceous mesoporous nanotubes (denoted as NC-MeNTs) were synthesized with the similar method without the addition of PA.

**Materials Characterization:** The morphologies and microstructures of the synthesized materials were characterized by field-emission scanning electron microscopy (FESEM, JSM-6700F, Japan), transmission electron microscopy (TEM, JOEL, JEM-2010; Talos F200X). X-ray diffraction (XRD, TTR-III, Japan) and Raman spectrometer (Renishaw inVia) were used to collected analyze the structural

information of the obtained samples. The surface chemical state of the obtained samples was performed by X-ray photoelectron spectroscopy (XPS, ESCALAB 250). The specific surface area and pore distribution were investigated via Brunauer-Emmett-Teller (BET; Tristar II 3020M).

**Electrochemical Measurement:** LIR2016 coin-type cells were assembled for all electrochemical measurements. The SIBs and KIBs used pure sodium or potassium foil as the counter electrodes and assembled in an argon-filled glove, a glass microfiber filters (Whatman, GF/F) as the separator, 1.0 M NaClO<sub>4</sub> in EC: DEC=1:1 Vol% with 5.0% FEC and 1.0 M KFSI in EMC=100 Vol% as the electrolytes, respectively. The active materials, carbon black (Super-P) and polyvinylidene difluoride (PVDF) binder (the weight ratio 70:20:10) were mixed in N-methyl-2-pyrrolidinone (NMP) solvent. The slurry coated on copper foil and dried in a vacuum oven at 100 °C for 12 h. The mass loadings of the anode materials are 0.8-1.0 mg cm<sup>-2</sup>. Galvanostatic charge/discharge measurements were tested on a Neware BTS-610 battery tester. Cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) were measured by a CHI 660B electrochemical workstation.

**Fabrication of Potassium ion Hybrid Capacitors:** For the Potassium-ion hybrid capacitors (KIHCs), the PNC-MeNT//AC KIHCs devices were assembled employing the preactivated PNC-MeNT anode and active carbon (AC) cathode with a mass ratio ranging from 1:1 to 1:3. The AC electrode was constructed by mixing AC, Super-P and PVDF (the weight ratio 70:20:10) on aluminum foils. The PNC-MeNT anode in K-half cells was cycled for 5 cycles at 0.1 A g<sup>-1</sup> and then assembled with AC cathode in the Ar-filled glove box. The energy/power density of the PNC-MeNT//AC KIHCs was calculated based on the following equations:<sup>1</sup>

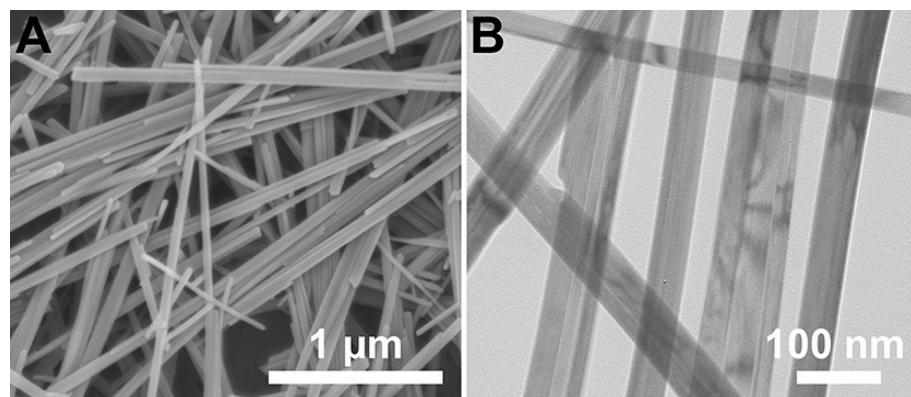
$$E = P \times t \quad (1)$$

$$P = V_{ave} \times i / m \quad (2)$$

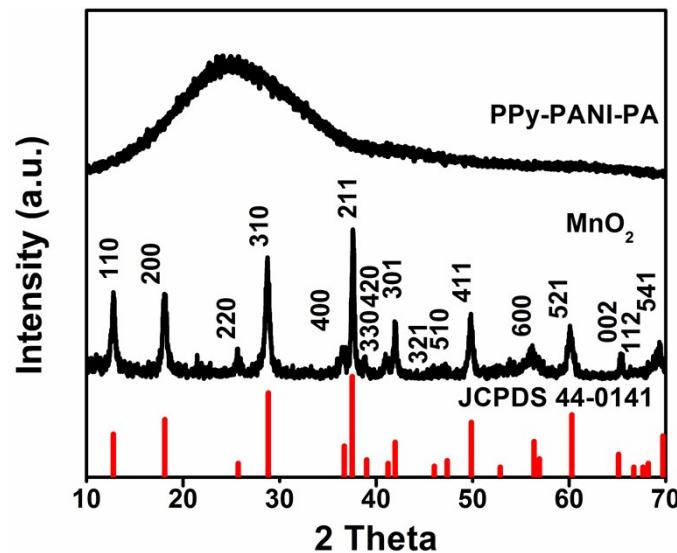
$$V_{ave} = (V_{max} + V_{min}) / 2 \quad (3)$$

Where  $t$  is the discharge time,  $i$  is the discharge current,  $m$  is the total mass of the active materials in both anode and cathode, and  $V_{max}$  is the potential at the beginning of discharge after the  $IR$  drop,  $V_{min}$  is voltage at the end of the discharge.

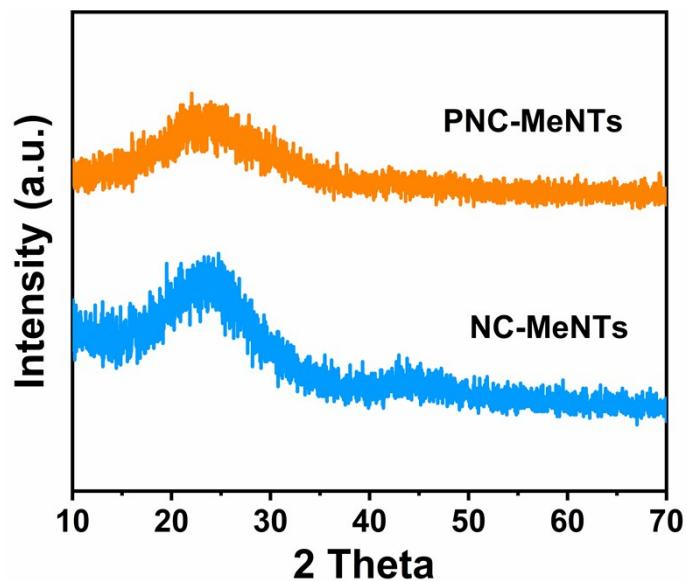
## 1. Characterization



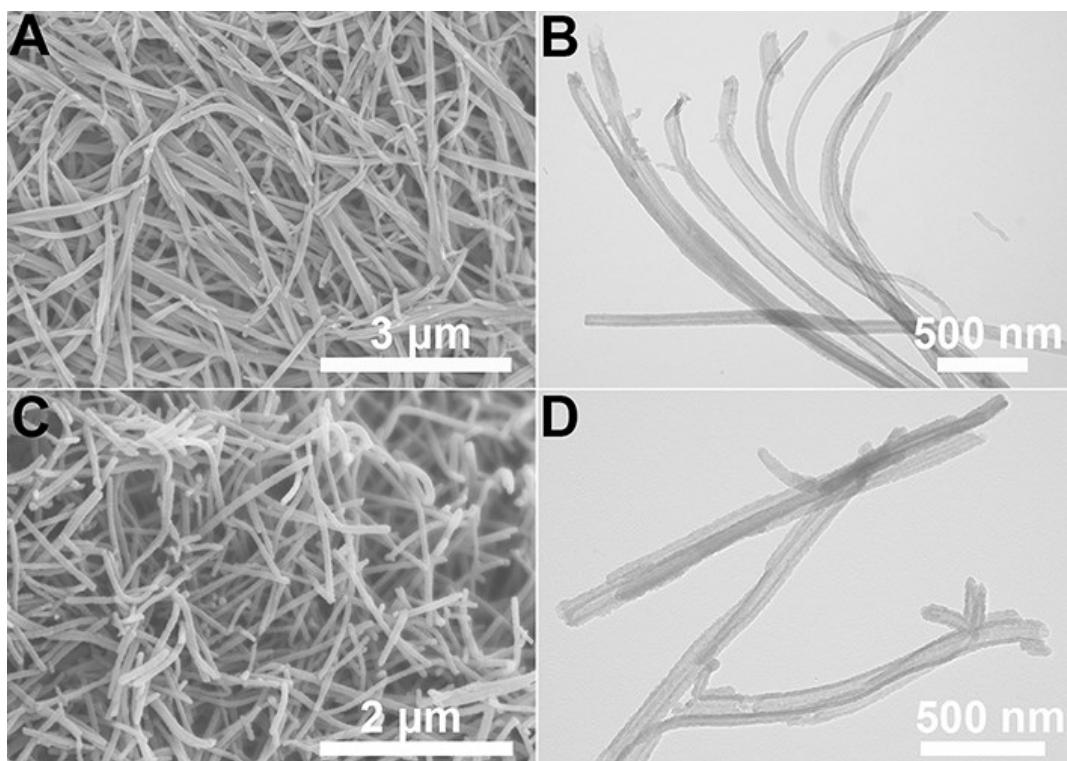
**Figure S1.** (A) The FESEM and (B) TEM images of the  $\text{MnO}_2$  nanowires.



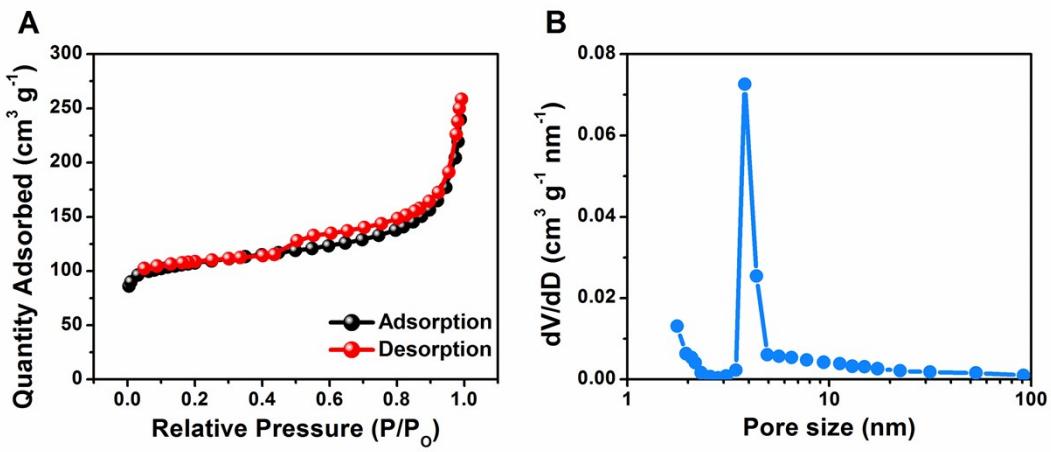
**Figure S2.** XRD patterns of  $\text{MnO}_2$  nanowires and PPy-PANI-PA composite nanotubes.



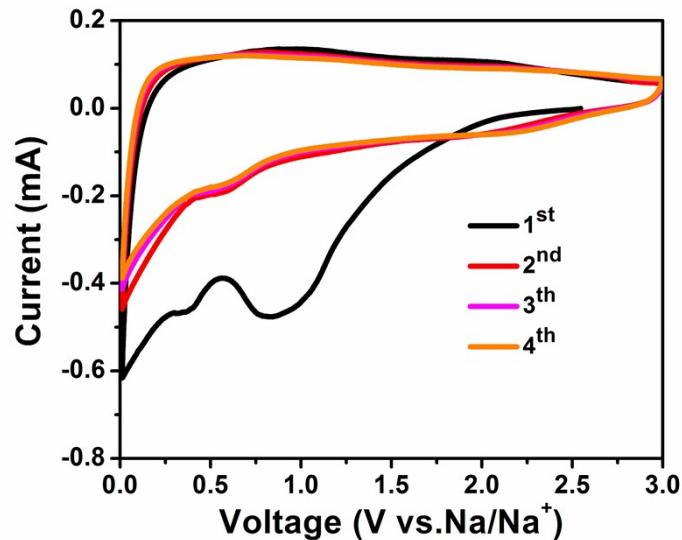
**Figure S3.** XRD patterns of PNC-MeNTs and NC-MeNTs.



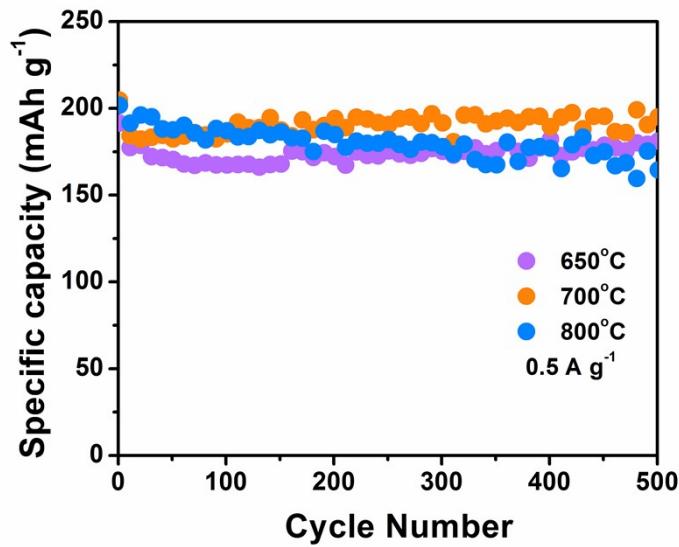
**Figure S4.** The SEM (A) and TEM (B) images of the NC-MeNTs precursor; the SEM (C) and (D) TEM images of the NC-MeNTs.



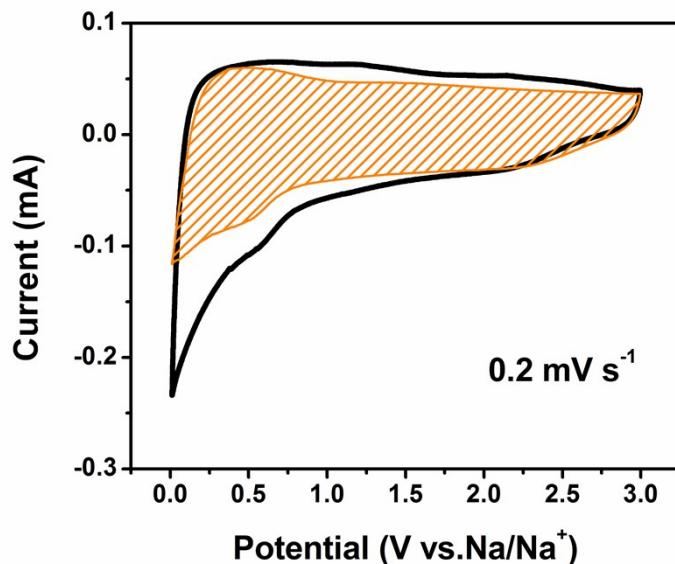
**Figure S5.**  $N_2$  adsorption/desorption isotherm (A) and pore-size distribution (B) of NC-MeNTs.



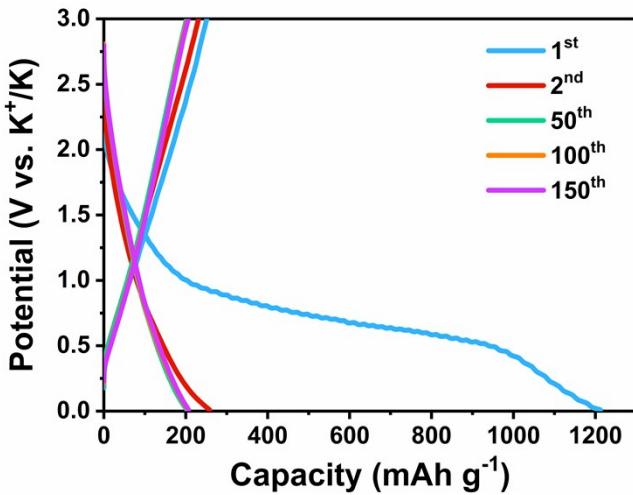
**Figure S6.** CV curves of PNC-MeNT anode at a scan rate of  $0.2 \text{ mV s}^{-1}$  for  $\text{Na}^+$  storage.



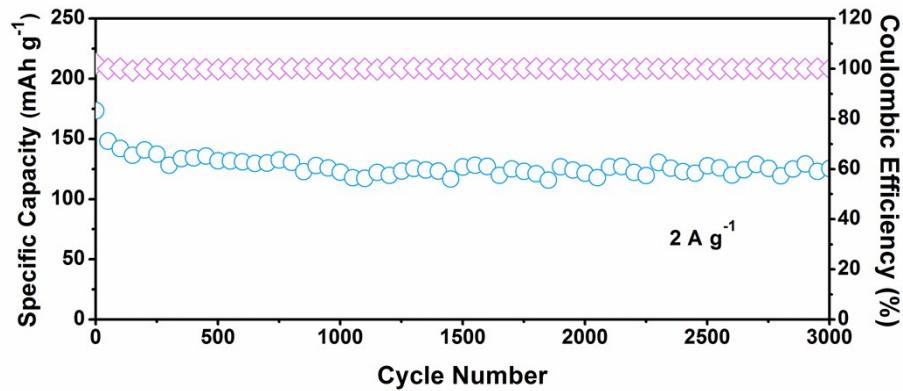
**Figure S7.** Cycling performance of PNC-MeNTs samples calcinated at different temperatures at a current density of  $0.5 \text{ A g}^{-1}$  for SIBs.



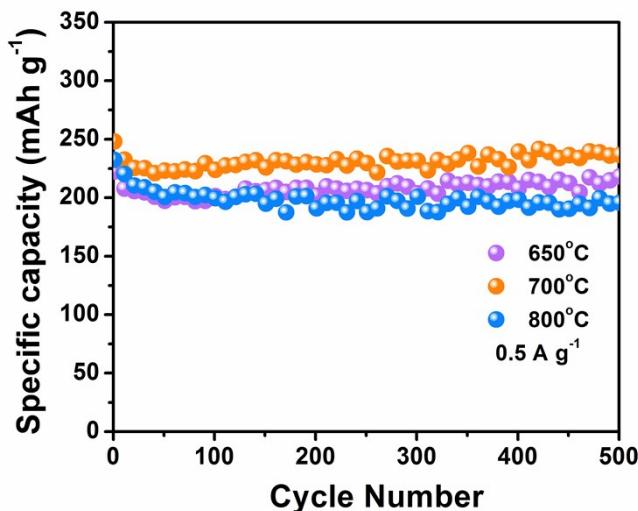
**Figure S8.** Capacitive contribution (shaded region) at a scan rate of  $0.2 \text{ mV s}^{-1}$  of PNC-MeNTs for SIBs.



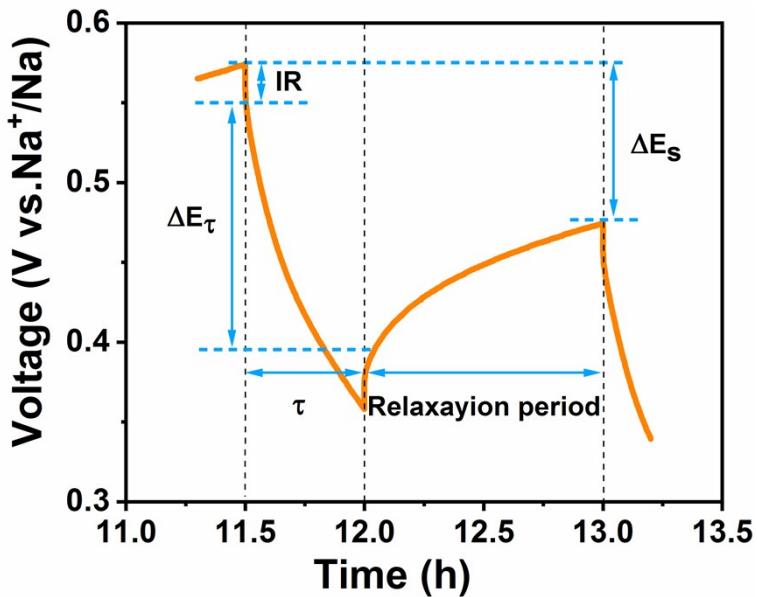
**Figure S9.** The charge/discharge profiles of PNC-MeNTs at  $0.1 \text{ A g}^{-1}$  for KIBs.



**Figure S10.** Long cycling performance of the PNC-MeNTs at a high current density of  $2 \text{ A g}^{-1}$  over 3000 cycles.



**Figure S11.** Cycling stability of PNC-MeNTs samples calcinated at different temperatures at  $0.5 \text{ A g}^{-1}$  for KIBs.

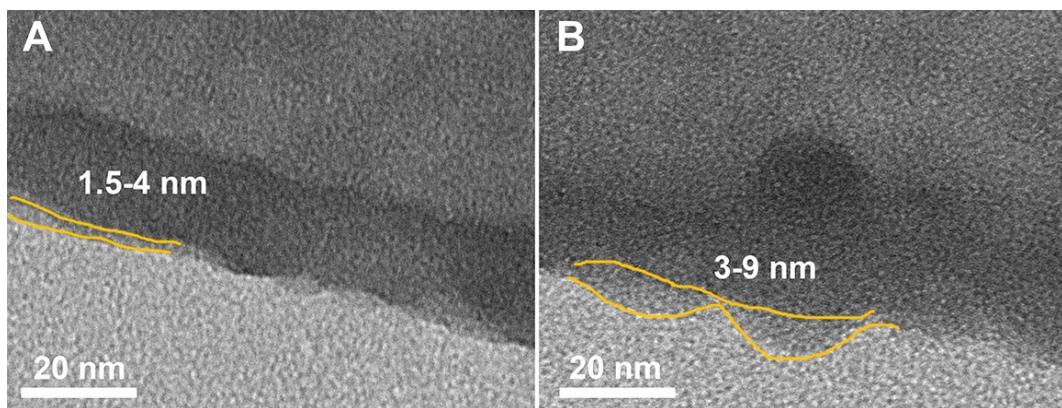


**Figure S12.**  $\Delta E_s$  and  $\Delta E_\tau$  profile of PNC-MeNTs during discharge process for SIBs.

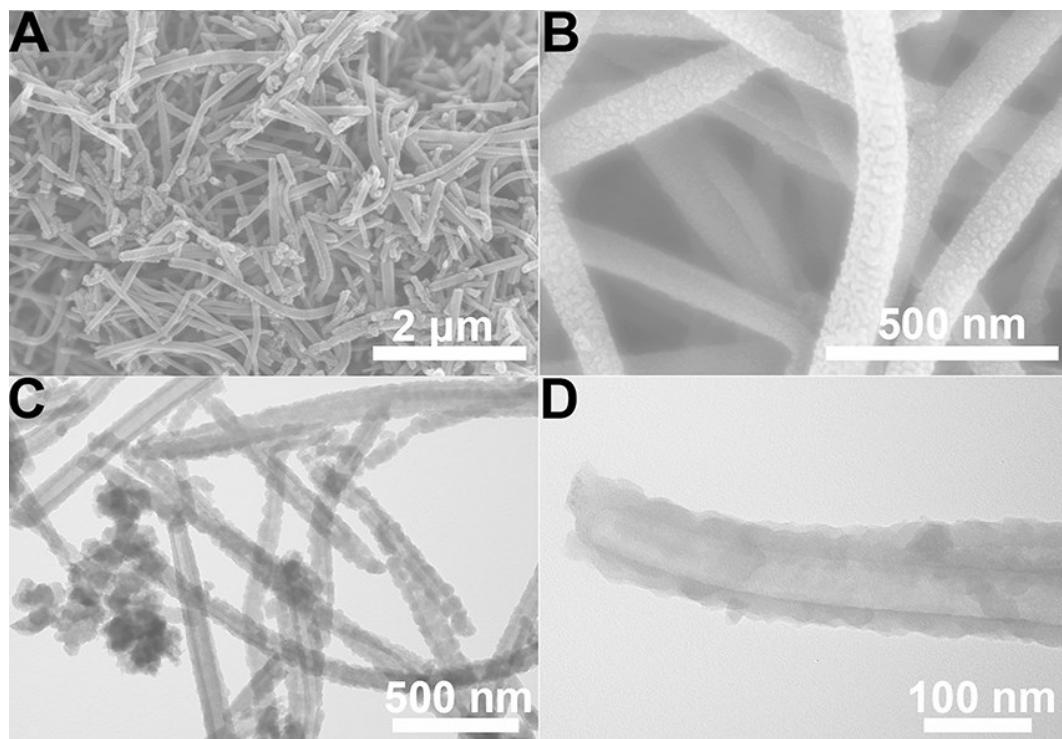
The galvanostatic intermittent titration (GITT) technique is employed to ascertain the effect on sodium/potassium ion diffusion kinetics with a pulse current at  $0.05 \text{ A g}^{-1}$  for 30 min between rest intervals for 60 min. The diffusion coefficient ( $D$ ) can be calculated from the following equation:<sup>2,3</sup>

$$D = \frac{4}{\pi\tau} \left( \frac{m_B V_M}{M_B S} \right)^2 \left( \frac{\Delta E_s}{\Delta E_\tau} \right)^2 \quad (4)$$

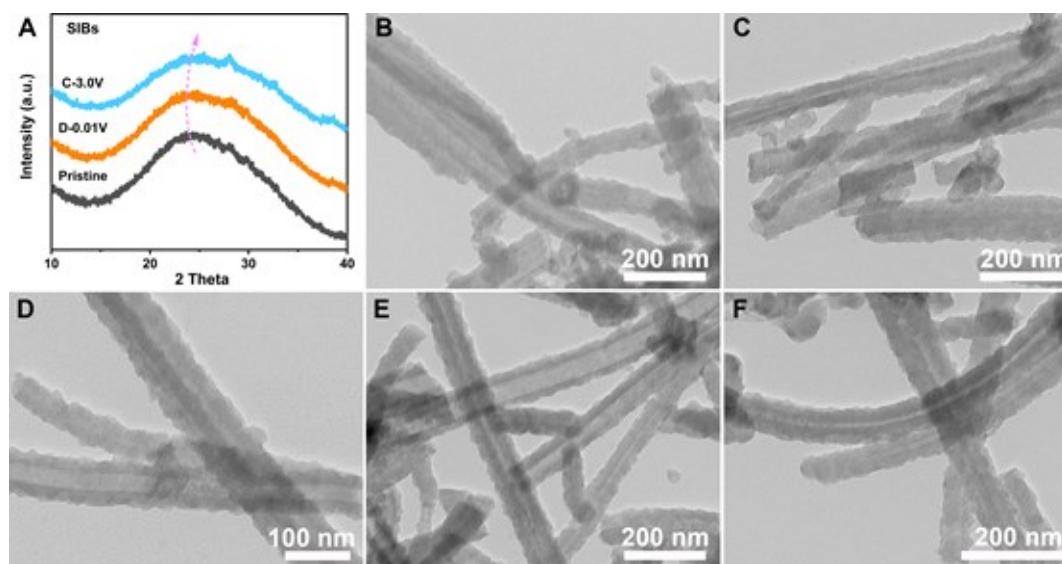
Where  $\tau$  is the time duration of the current pulse;  $m_B$  is the mass loading of the active material;  $V_M$  is the molar volume of the electrode;  $M_B$  is the molar mass of electrode material;  $S$  is the area of the electrode;  $\Delta E_s$  is the steady-state voltage change between before and after the current pulse;  $\Delta E_\tau$  is the voltage change during the current pulse.



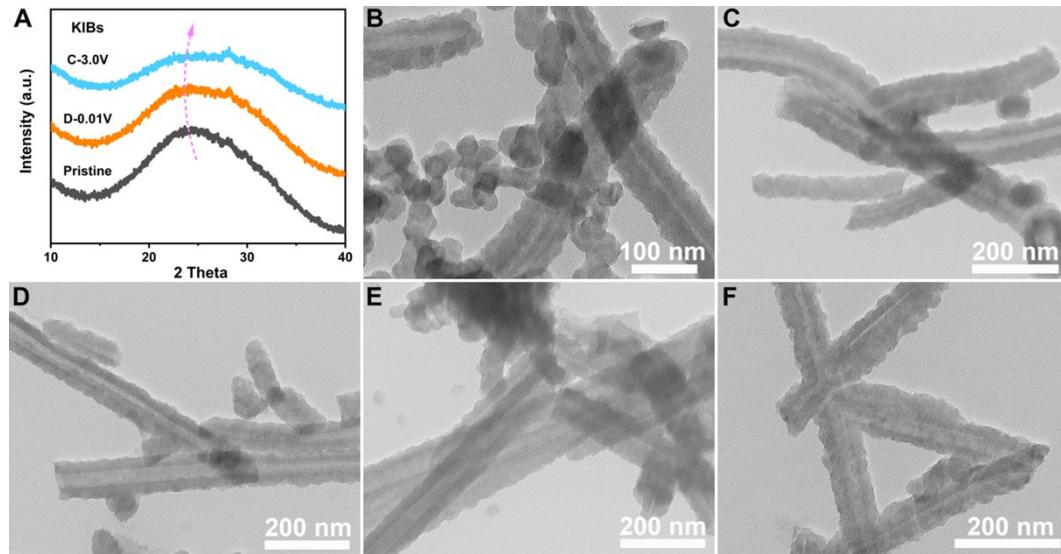
**Figure S13.** The TEM images of PNC-MeNTs after discharged to 0.01 V at  $0.01 \text{ A g}^{-1}$  in SIBs (A) and KIBs (B).



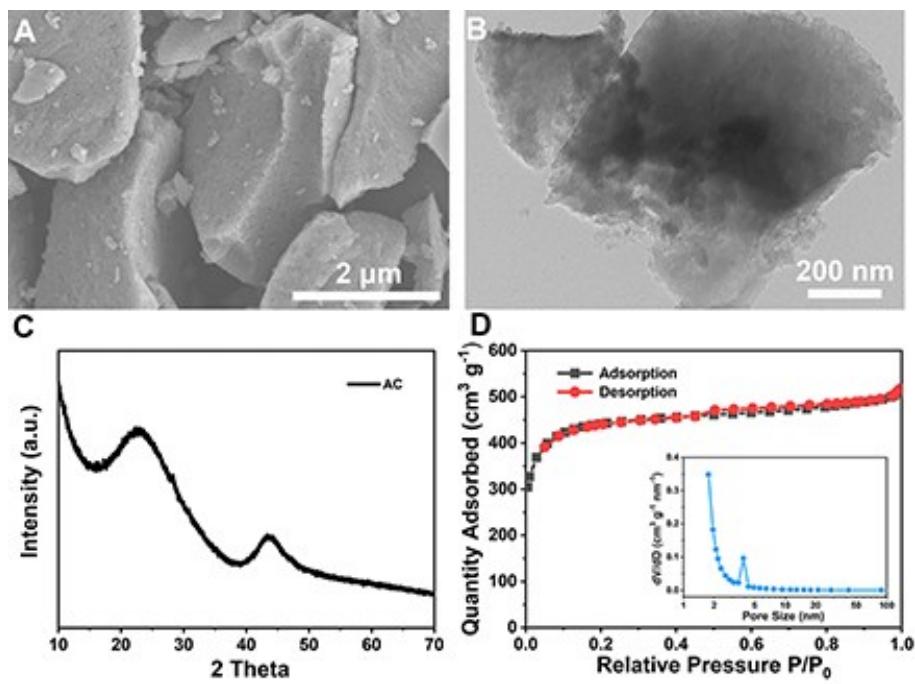
**Figure S14.** The SEM (A, B) and TEM (C, D) images of PNC-MeNTs after 200 cycles at  $0.1 \text{ A g}^{-1}$  for SIBs.



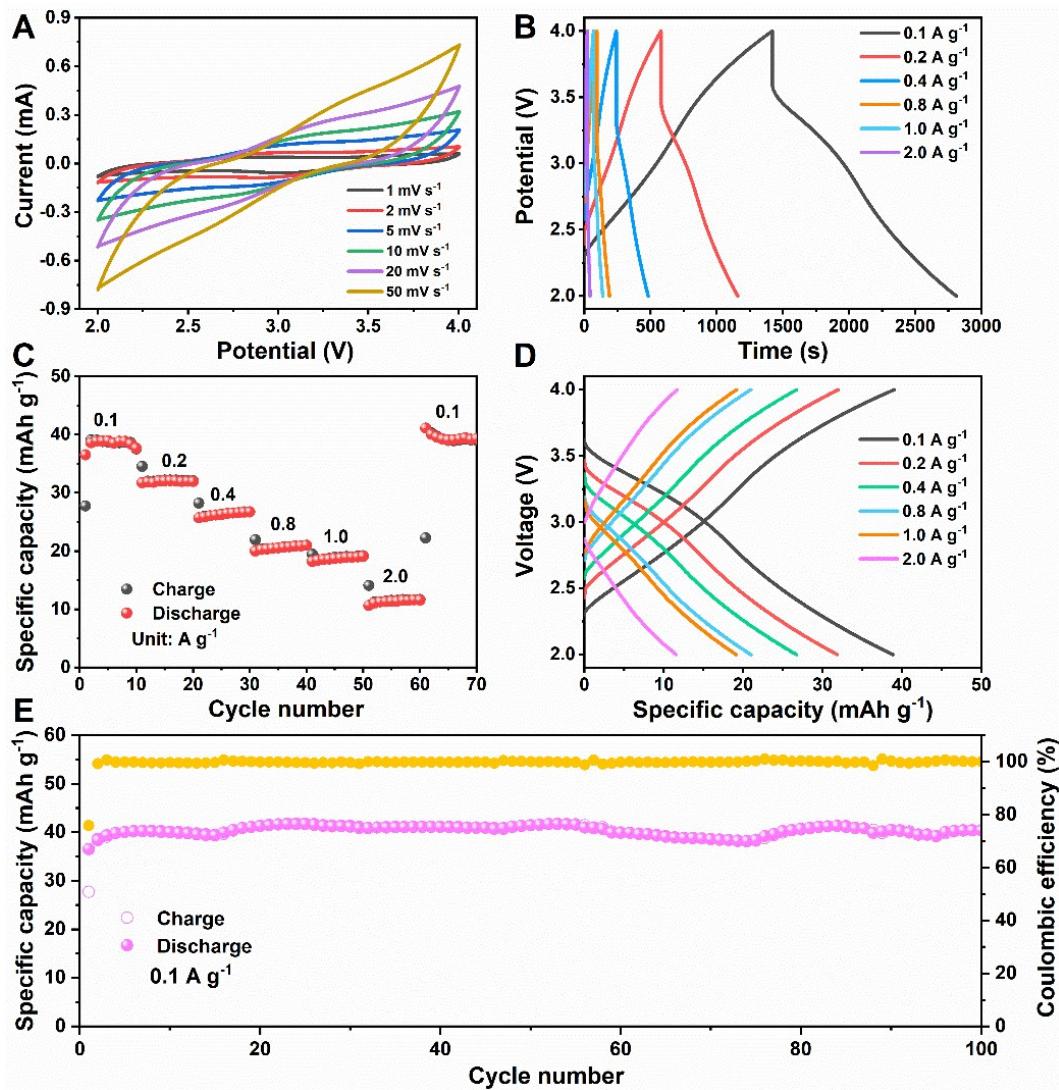
**Figure S15.** The ex situ XRD patterns (A); The TEM images of PNC-MeNTs in different sodiation/desodiation states at a current density of  $0.1 \text{ A g}^{-1}$ : the initial state(B), discharged to  $1.5 \text{ V}$  (C) and  $0.01 \text{ V}$  (D), charged to  $1.5 \text{ V}$  (E) and  $3.0 \text{ V}$  (F).



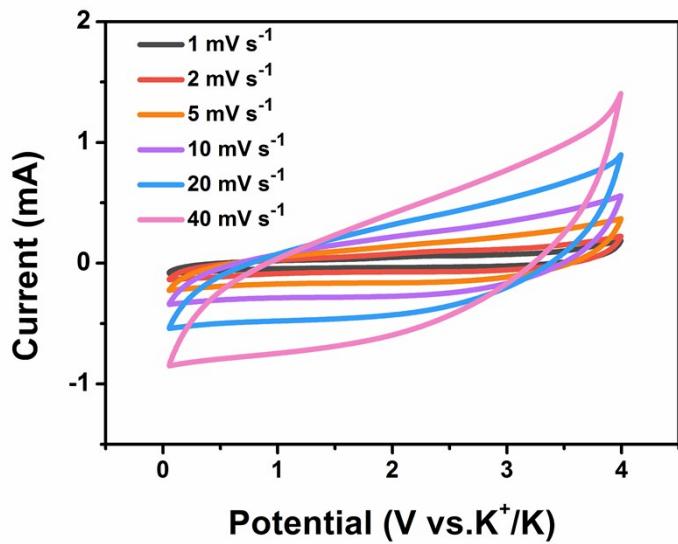
**Figure S16.** The ex situ XRD patterns (A); The TEM images of PNC-MeNTs in different potassiation/depotassiation states at a current density of  $0.1 \text{ A g}^{-1}$ : the initial state(B), discharged to  $1.5 \text{ V}$  (C) and  $0.01 \text{ V}$  (D), charged to  $1.5 \text{ V}$  (E) and  $3.0 \text{ V}$  (F).



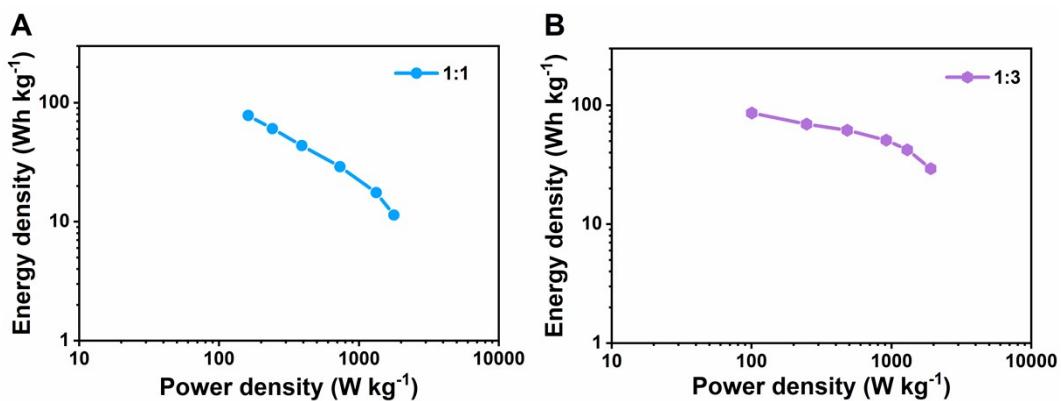
**Figure S17.** (A) The SEM; (B) TEM images; (C) XRD pattern and (D)  $\text{N}_2$  adsorption/desorption isotherm (inset: pore-size distribution) for commercial AC.



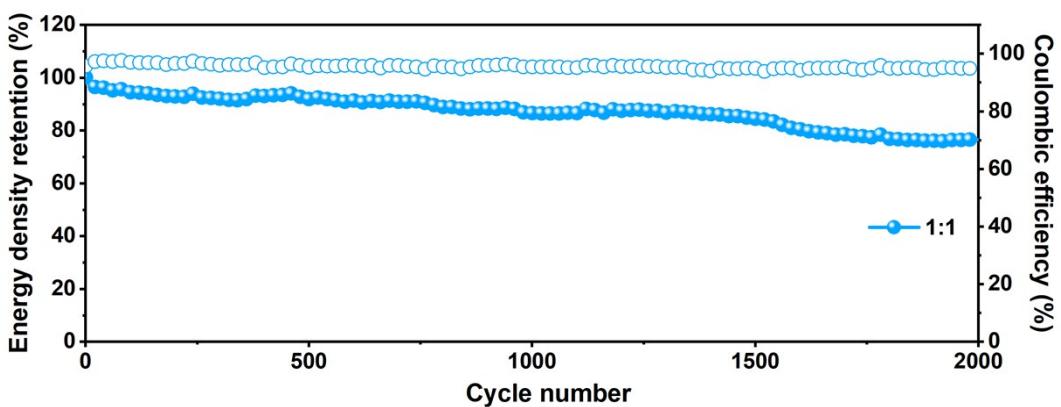
**Figure S18.** (A) CV curves at various scan rates and GCD curves at various current densities of AC electrode in 2.0-4.0 V (B); (C) Rate performance and (E) charge/discharge curves at different current densities of AC; (F) Cycling stability of AC at a current density of 0.1 A g<sup>-1</sup> in half cell.



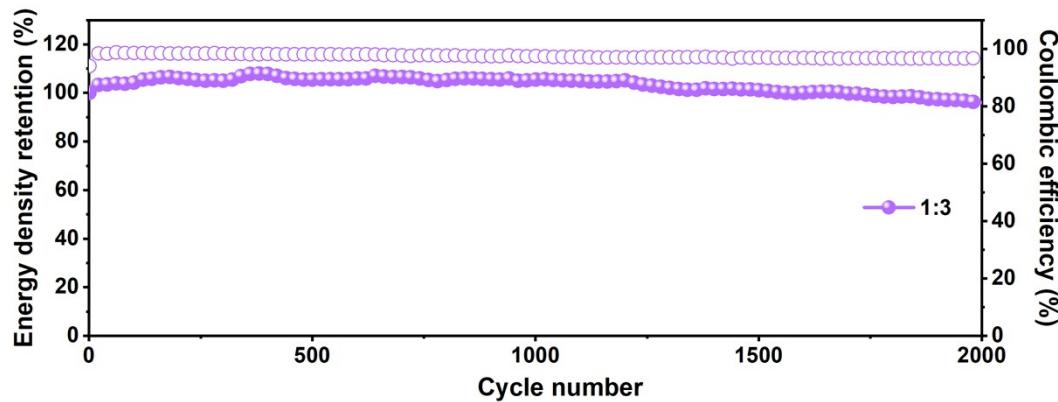
**Figure S19.** CV curves at different scan rates of the PNC-MeNT//AC KIHCs with the mass ratio of 1:1.5.



**Figure S20.** Ragone plot of the PNC-MeNT//AC KIHCs with mass ratios of 1:1 (A) and 1:3 (B).



**Figure S21.** Cycling performance of the PNC-MeNT//AC KIHCs with the mass ratio of 1:1 at 0.8 A g<sup>-1</sup>.



**Figure S22.** Cycling stability of the PNC-MeNT//AC KIHCs with the mass ratio of 1:3 at 0.8 A g<sup>-1</sup>.

**Table S1.** Elemental analysis of PNC-MeNTs and NC-MeNTs.

Atomic content [%]	C	N	P
PNC-MeNTs	81.39	15.03	3.58
NC-MeNTs	87.52	12.48	---

**Table S2.** Comparison of the electrochemical performances of reported carbon-based carbon materials for SIBs.

Materials	Rate capability (mAh g <sup>-1</sup> )	Cyclability (mAh g <sup>-1</sup> )	Reference
P-CNSs	269 at 0.2 A g <sup>-1</sup>		
	235 at 0.5 A g <sup>-1</sup>		
	208 at 1 A g <sup>-1</sup>	196.9/1000 cycle/1 A g <sup>-1</sup>	Ref 4
	169 at 2 A g <sup>-1</sup>	108.8/5000 cycle/5 A g <sup>-1</sup>	
	143 at 5 A g <sup>-1</sup>		
N, P-doped carbon sheets	117 at 10 A g <sup>-1</sup>		
	233 at 0.1 A g <sup>-1</sup>		
	204 at 0.2 A g <sup>-1</sup>	202/200 cycle/0.2 A g <sup>-1</sup>	Ref 5
	177 at 0.5 A g <sup>-1</sup>	103/2000 cycle/1 A g <sup>-1</sup>	
	143 at 1 A g <sup>-1</sup>		
3D PCFs	122 at 2 A g <sup>-1</sup>		
	290 at 0.2 A g <sup>-1</sup>		
	253 at 0.5 A g <sup>-1</sup>		
	200 at 1 A g <sup>-1</sup>	99.8/10 000 cycle/5 A g <sup>-1</sup>	Ref 6
	166 at 2 A g <sup>-1</sup>		
SC-NS	130 at 5 A g <sup>-1</sup>		
	104 at 10 A g <sup>-1</sup>		
	232.2 at 0.02 A g <sup>-1</sup>	128.7/3500 cycle/0.8 A g <sup>-1</sup>	Ref 7
	103.8 at 1 A g <sup>-1</sup>		
3DFC-700	278 at 0.1 A g <sup>-1</sup>	175/1000 cycle/0.5 A g <sup>-1</sup>	Ref 8
	227 at 0.2 A g <sup>-1</sup>	99/10 000 cycle/10 A g <sup>-1</sup>	
	199 at 0.5 A g <sup>-1</sup>		

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	176 at 2 A g <sup>-1</sup>		
	127 at 10 A g <sup>-1</sup>		
	264 at 0.1 A g <sup>-1</sup>		
	231 at 0.2 A g <sup>-1</sup>		
	162 at 0.5 A g <sup>-1</sup>		
huCP/g-C <sub>3</sub> N <sub>4</sub>	134 at 1 A g <sup>-1</sup>	110/4000 cycle/1 A g <sup>-1</sup>	Ref 9
	116 at 2 A g <sup>-1</sup>		
	93 at 5 A g <sup>-1</sup>		
	64 at 10 A g <sup>-1</sup>		
PCS	317 at 0.1 A g <sup>-1</sup>		
	204 at 0.2 A g <sup>-1</sup>		
	151 at 0.5 A g <sup>-1</sup>	240/200 cycle/0.1 A g <sup>-1</sup>	Ref 10
	116 at 1 A g <sup>-1</sup>		
	91 at 2 A g <sup>-1</sup>		
	70 at 5 A g <sup>-1</sup>		
FP-MP 5:2 1000	281 at 0.03 A g <sup>-1</sup>		
	259 at 0.06 A g <sup>-1</sup>		
	227 at 0.15 A g <sup>-1</sup>		
	174 at 0.3 A g <sup>-1</sup>	191/150 cycle/0.15 A g <sup>-1</sup>	Ref 11
	121 at 0.6 A g <sup>-1</sup>		
	78 at 1.2 A g <sup>-1</sup>		
Carbon derived from sweet gum	339.1 at 0.05 A g <sup>-1</sup>		
	281.7 at 0.1 A g <sup>-1</sup>		
	237.4 at 0.2 A g <sup>-1</sup>	136.1/1000 cycle/1 A g <sup>-1</sup>	Ref 12
	169.6 at 0.5 A g <sup>-1</sup>		
	138.3 at 1 A g <sup>-1</sup>		
PNC-MeNTs	<b>283.5 at 0.1 A g<sup>-1</sup></b>		
	<b>239.0 at 0.2 A g<sup>-1</sup></b>	<b>256.2/200 cycle/0.1 A g<sup>-1</sup></b>	
	<b>216.2 at 0.5 A g<sup>-1</sup></b>	<b>214.1/1000 cycle/1 A g<sup>-1</sup></b>	<b>This work</b>
	<b>192.5 at 1 A g<sup>-1</sup></b>	<b>94.7/10 000 cycle/10 A g<sup>-1</sup></b>	
	<b>174.1 at 2 A g<sup>-1</sup></b>		

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**147.3 at 5 A g<sup>-1</sup>**  
**123.3 at 10 A g<sup>-1</sup>**

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**Table S3.** Comparison of the electrochemical performances of reported carbon-based carbon materials for KIBs.

Materials	Rate capability (mAh g <sup>-1</sup> )	Cyclability (mAh g <sup>-1</sup> )	Reference
OMC	286.1 at 0.05 A g <sup>-1</sup>		
	255.1 at 0.1 A g <sup>-1</sup>		
	218.8 at 0.2 A g <sup>-1</sup>	146.5/1000 cycle/1 A g <sup>-1</sup>	Ref 13
	186.3 at 0.5 A g <sup>-1</sup>		
	144.2 at 1 A g <sup>-1</sup>		
S/N@C	320 at 0.05 A g <sup>-1</sup>		
	235 at 0.1 A g <sup>-1</sup>		
	160.2 at 0.5 A g <sup>-1</sup>	200/400 cycle/0.1 A g <sup>-1</sup>	Ref 14
	123.5 at 1 A g <sup>-1</sup>	65/900 cycle/1 A g <sup>-1</sup>	
	91.2 at 2 A g <sup>-1</sup>		
PNCM	388 at 0.05 A g <sup>-1</sup>		
	319 at 0.1 A g <sup>-1</sup>		
	286 at 0.2 A g <sup>-1</sup>		
	253 at 0.5 A g <sup>-1</sup>	152/3000 cycle/1 A g <sup>-1</sup>	Ref 15
	225 at 1 A g <sup>-1</sup>		
SC-NS	199 at 2 A g <sup>-1</sup>		
	247 at 0.1 A g <sup>-1</sup>		
	185 at 0.2 A g <sup>-1</sup>		
	162 at 0.5 A g <sup>-1</sup>	117.2/3000 cycle/1 A g <sup>-1</sup>	Ref 16
	146 at 1 A g <sup>-1</sup>		
3DFC-700	130 at 2 A g <sup>-1</sup>		
	340 at 28 mA g <sup>-1</sup>	250/150 cycle/0.14 A g <sup>-1</sup>	
	301 at 56 mA g <sup>-1</sup>	~153/500 cycle/0.28 A g <sup>-1</sup>	Ref 17

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	253 at 140 mA g <sup>-1</sup>		
	204 at 280 mA g <sup>-1</sup>		
	117 at 560 mA g <sup>-1</sup>		
	354 at 0.05 A g <sup>-1</sup>		
	317 at 0.1 A g <sup>-1</sup>		
MCOs	223 at 0.2 A g <sup>-1</sup>	100/1300 cycle/1 A g <sup>-1</sup>	Ref 18
	178 at 0.5 A g <sup>-1</sup>	80/3000 cycle/2 A g <sup>-1</sup>	
	110 at 1 A g <sup>-1</sup>		
	245 at 0.05 A g <sup>-1</sup>		
	194 at 0.1 A g <sup>-1</sup>		
OLC	167 at 0.2 A g <sup>-1</sup>	151/1000 cycle/0.5 A g <sup>-1</sup>	Ref 19
	140 at 0.5 A g <sup>-1</sup>	111/1000 cycle/2 A g <sup>-1</sup>	
	121 at 1 A g <sup>-1</sup>		
	106 at 2 A g <sup>-1</sup>		
	326 at 0.05 A g <sup>-1</sup>		
	252 at 0.1 A g <sup>-1</sup>		
N-HCNs	210 at 0.2 A g <sup>-1</sup>	201/100 cycle/0.1 A g <sup>-1</sup>	Ref 20
	182 at 0.5 A g <sup>-1</sup>	154/2500 cycle/1 A g <sup>-1</sup>	
	162 at 1 A g <sup>-1</sup>		
	141 at 2 A g <sup>-1</sup>		
NOGCN	476 at 0.05 A g <sup>-1</sup>		
	114 at 1 A g <sup>-1</sup>	131/300 cycle/0.5 A g <sup>-1</sup>	Ref 21
	<b>258.6 at 0.1 A g<sup>-1</sup></b>		
	<b>206.6 at 0.2 A g<sup>-1</sup></b>	<b>210.2/150 cycle/0.1 A g<sup>-1</sup></b>	
PNC-MeNTs	<b>169.6 at 0.5 A g<sup>-1</sup></b>	<b>188.7/3000 cycle/1 A g<sup>-1</sup></b>	This work
	<b>146.1 at 1 A g<sup>-1</sup></b>	<b>125.5/3000 cycle/2 A g<sup>-1</sup></b>	
	<b>106.9 at 2 A g<sup>-1</sup></b>		

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**Table S4.** Comparison of the electrochemical performances of the PNC-MeNT//AC KIHCs with recent literatures about potassium ion hybrid capacitors.

Materials	Voltage window (V)	Energy density (Wh kg <sup>-1</sup> ) /Power density (W kg <sup>-1</sup> )	Cycling life	Reference
NHCS//ANHCS PIHC	0.01-4.0	114.2/100.5 19.1/8203	93%/2000/0.5 A g <sup>-1</sup> 80.4%/5000/2 A g <sup>-1</sup>	Ref 22
CTP@C//AC KIC	1.0-4.0	80/32 34/5144	75.9%/4000/5 A g <sup>-1</sup>	Ref 23
PB//AC KIC	0-1.9	28/214 10.5/1890	98%/1200/2 A g <sup>-1</sup>	Ref 24
KTO//NGC KIC	0-3.5	58.2/160 11.8/7200	75.5%/5000/1 A g <sup>-1</sup>	Ref 25
graphite//AC KIC	0.5-3.5	12/22 11/1500	97%/55000	Ref 26
Co <sub>2</sub> P@rGO//AC KIC	1.0-4.0	87/12 10/4264.7	68%/1000/1 A g <sup>-1</sup>	Ref 27
FeSe <sub>2</sub> /AC PIHC	0.1-3.8	230/198 30/920	/	Ref 28
CFMS//CFMS PDIC	0.01-3.8	58/1158.2 39/7800	90%/10 000/2 A g <sup>-1</sup>	Ref 29
NCNTs//AC KIHC	0.01-4	117.1/112.8 25.7/1713.4	81.6%/2000/1 A g <sup>-1</sup>	Ref 30
SC//AC KIHC	0-4	120/96 13.3/599	97.5%/1500/0.75 A g <sup>-1</sup>	Ref 31
N,P-doped C//ADPC	1-4.2	72/257	75.6%/2000/1 A g <sup>-1</sup>	Ref 32

KIHC		18.5/5220		
NPG//AC	1-4	104.4/760.6	~65%/1000/1 A g <sup>-1</sup>	Ref 33
KIHC		41.6/14976		
HC//AC	1.5-4.2	77/2830	84%/50/0.4 A g <sup>-1</sup>	Ref 34
KIHC		4.5/5000		
S-N-PCNs//AC	0.01-4.0	187/99	86.4%/3000/1 A g <sup>-1</sup>	Ref 35
KIHC		76/5136		
<b>PNC-MeNT//AC</b>	<b>0.05-4.0</b>	<b>175.1@160.6</b>	<b>85.8%/3000/0.8 A g<sup>-1</sup></b>	<b>This work</b>
<b>KIHC</b>		<b>31.6@3034</b>		

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