Electronic Supplementary Material (ESI) for Energy & Environmental Science.

Supporting Information

Freestanding Film Made by Necklace-like N-doped Hollow Carbon with Hierarchical Pores for High-performance Potassium-Ion Storage

*Wenxiu Yang[a]†, Jinhui Zhou[a]†, Shuo Wang[a], Weiyu Zhang[a], Zichen Wang[a], Fan Lv[a], Kai Wang[a], Qiang Sun[a], and Shaojun Guo[a, b, c] **

[a] Department of Materials Science & Engineering, College of Engineering, Peking University, Beijing, 100871, China. E-mail: guosj@pku.edu.cn

[b] BIC-ESAT, College of Engineering, Peking University, Beijing, 100871, China.

[c] Department of Energy and Resources Engineering, College of Engineering, Peking University, Beijing, 100871, China.

†These authors contributed equally to this work.

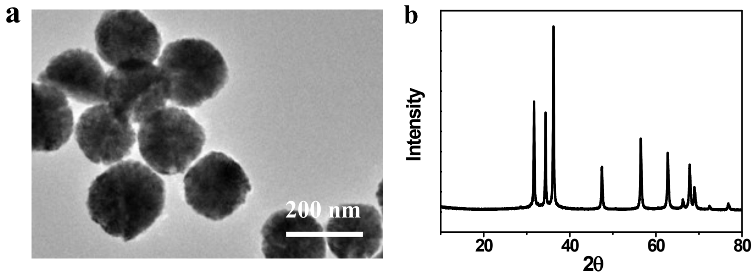

Figure S1 TEM image and XRD pattern of the ZnO nanospheres.

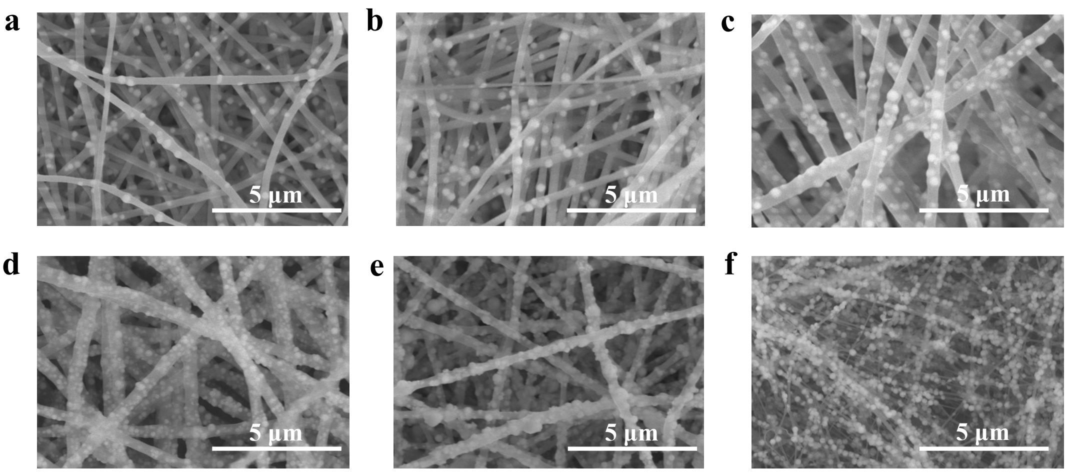

Figure S2 SEM images of *u*-$NHC_{x/y}$ made with different precursor proportions (ZnO:PAN): (a) 1:3, (b) 1:2, (c) 2:3, (d) 1:1, (e) 2:1 and (f) 3:1.

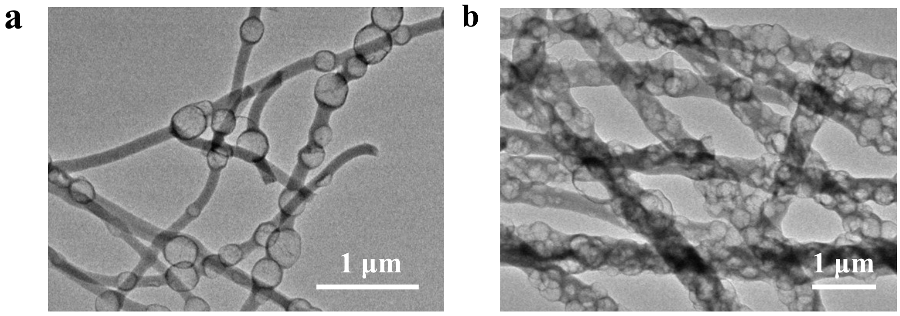

Figure S3 TEM images of (a) $NHC_{1/2}$-NH_3/Ar and (b) NHC_1-NH_3/Ar.

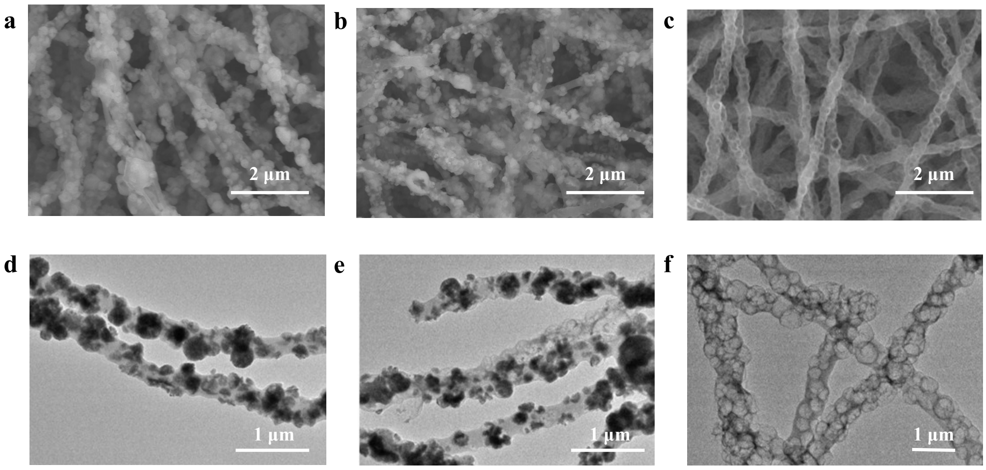

Figure S4 (a-c) SEM and (d-f) TEM images of NHC pyrolyzed at different temperature in Ar atmosphere: (a, d) NHC_2-Ar-600, (b, e) NHC_2-Ar-700 and (c, f) NHC_2-Ar-800.

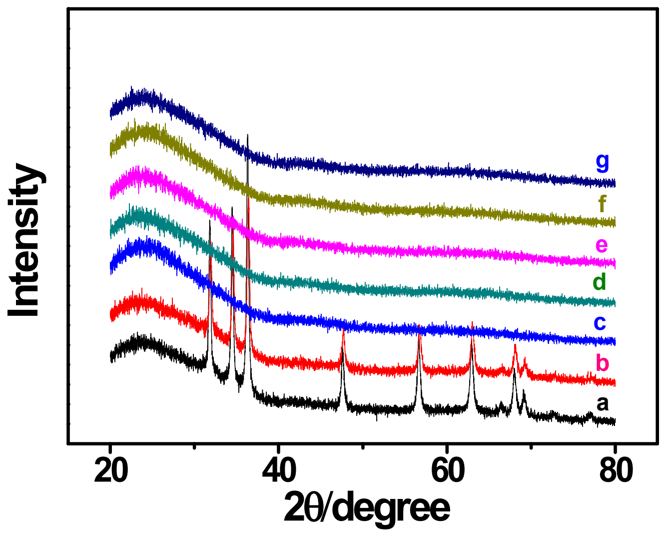

Figure S5 XRD patterns of the NHC materials: a (NHC_2-Ar-600), b (NHC_2-Ar-700), c (NHC_2-Ar-800), d (NHC_2-Ar-900), e ($NHC_{1/2}$-NH_3/Ar), e (NHC_1-NH_3/Ar) and g (NHC_2-NH_3/Ar).

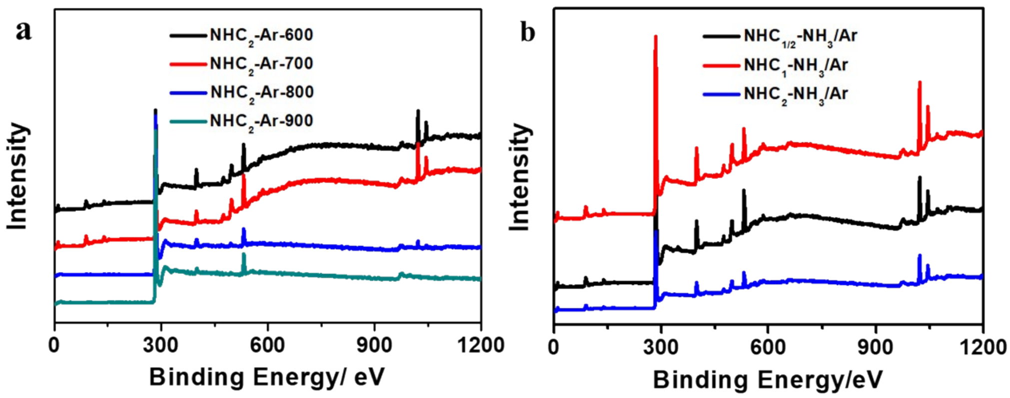

Figure S6 XPS spectra of different NHC materials.

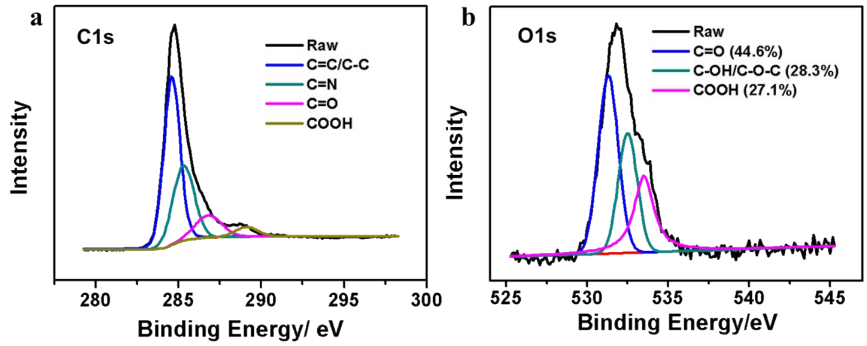

Figure S7 High-resolution XPS spectra of NHC_2-NH_3/Ar. (a) C_{1s} and (b) O_{1s}.

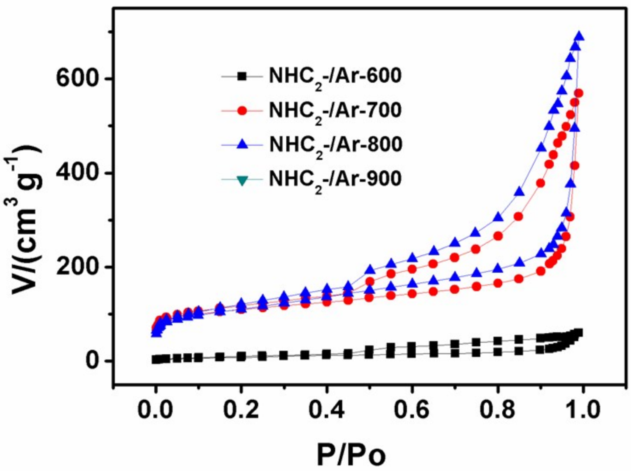

Figure S8 Nitrogen adsorption-desorption isotherms of the NHC materials.

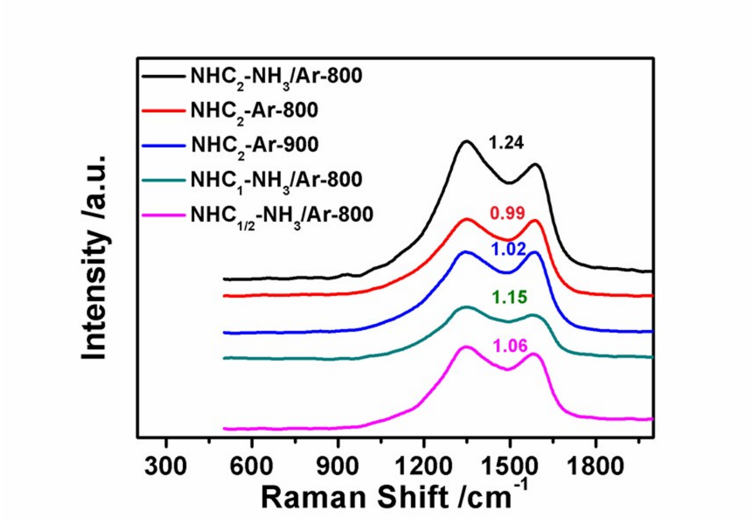

Figure S9 Raman spetra of the NHC materials.

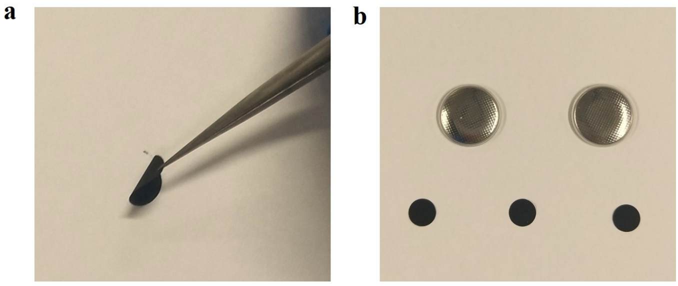

Figure S10 (a, b) Photographs of the NHC_2-NH_3/Ar.

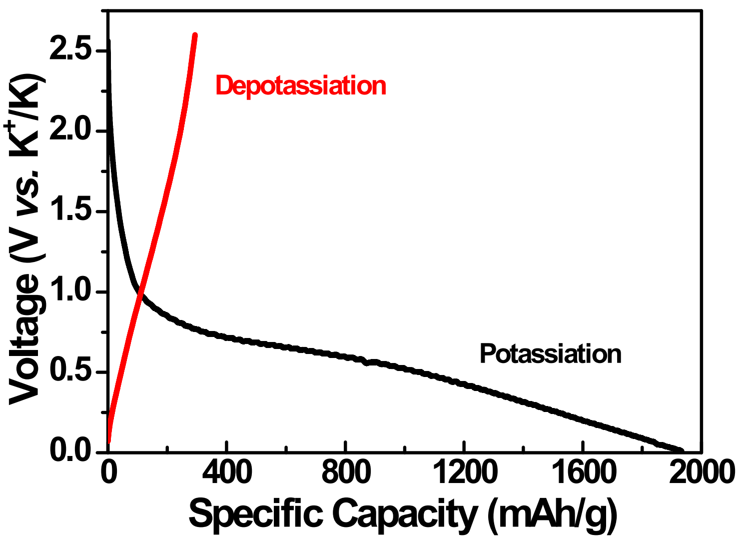

Figure S11. Potassiation and depotassiation profiles of the NHC_2-NH_3/Ar electrode for the first cycle at 100 mA/g.

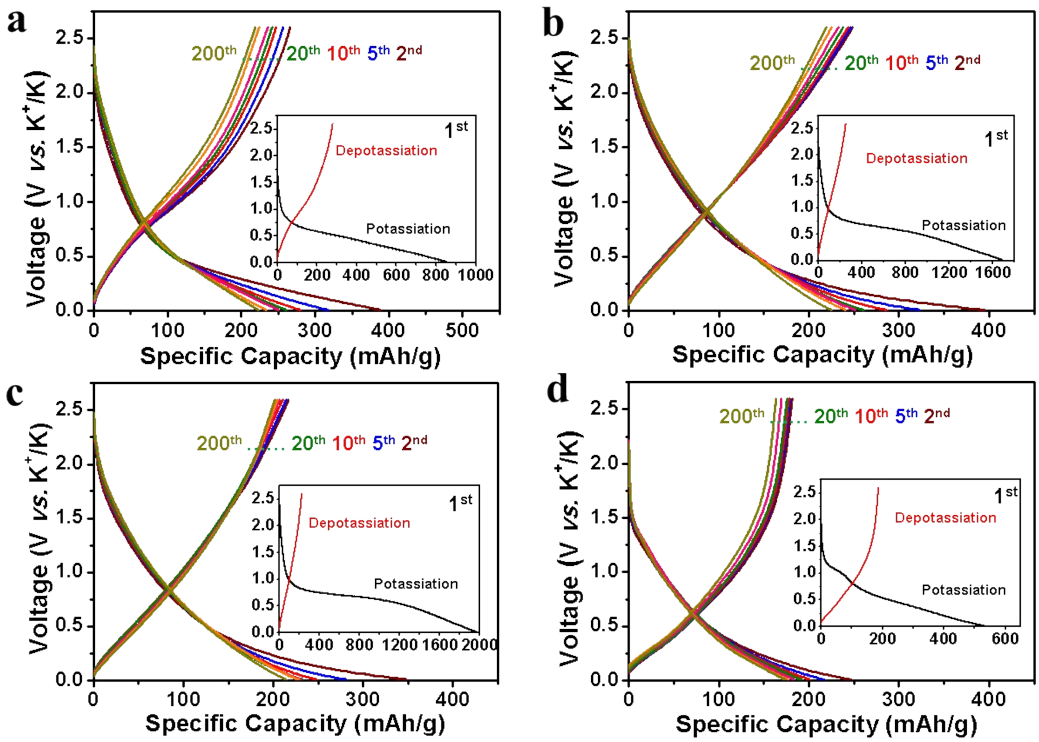

Figure S12. The 2nd, 5th, 10th, 20th, 50th, 100th and 200th potassiation and depotassiation profiles of the series of NHCFs materials at the current density of 100 mA/g: (a) NHC_1-NH_3/Ar, (b) NHC_2-Ar-800, (c) NHC_2-Ar-900 and (d) PAN-NH_3/Ar.

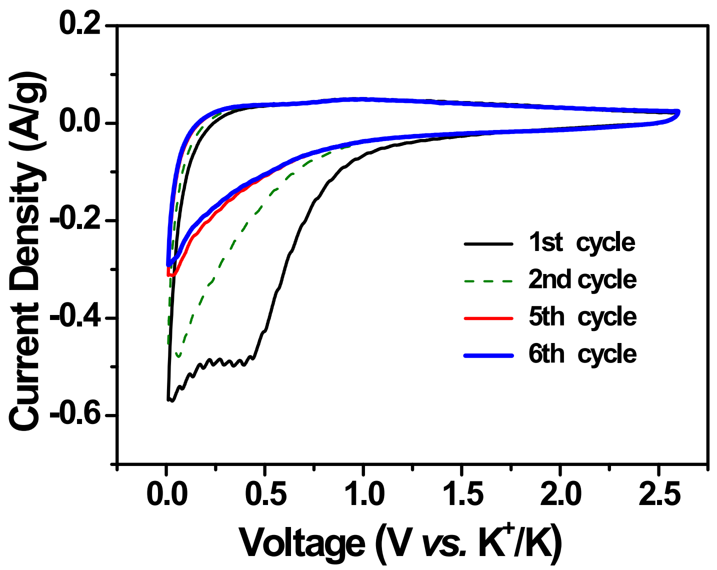

Figure S13. Cyclic voltammetry (CV) of the NHC_2-NH_3/Ar electrode for PIBs between 0.01 V and 2.5 V with a scan rate of 0.1 mV/s.

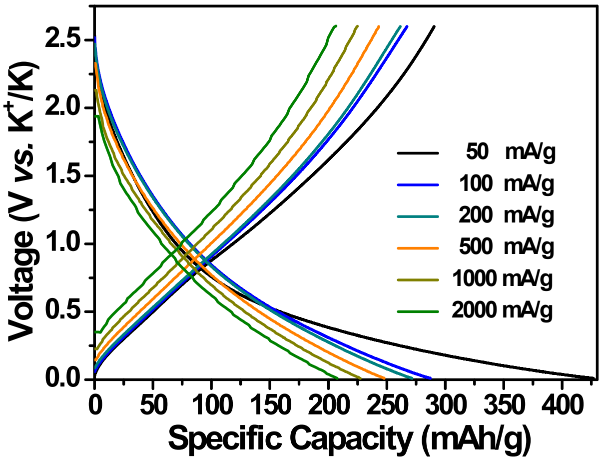

Figure S14. The potassiation and depotassiation profiles of the NHC_2-NH_3/Ar electrode at different current densities from 50 to 2000 mA/g.

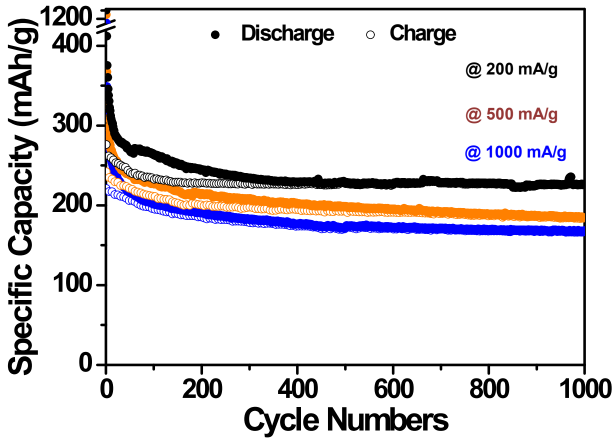

Figure S15.The cycles of the NHC_2-NH_3/Ar electrode measured under different current densities of 200, 500 and 1000 mA/g.

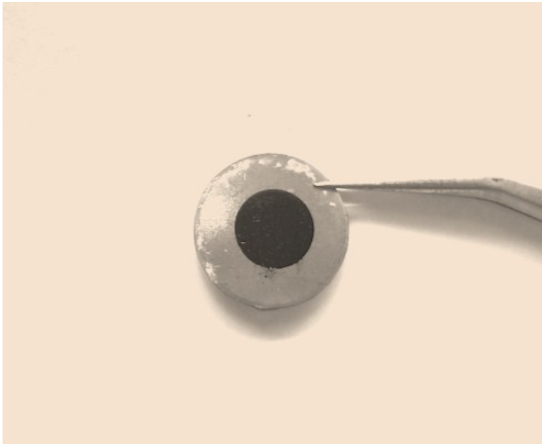

Figure S16. Digital photograph of NHC_2-NH_3/Ar electrode after 1000 cycles

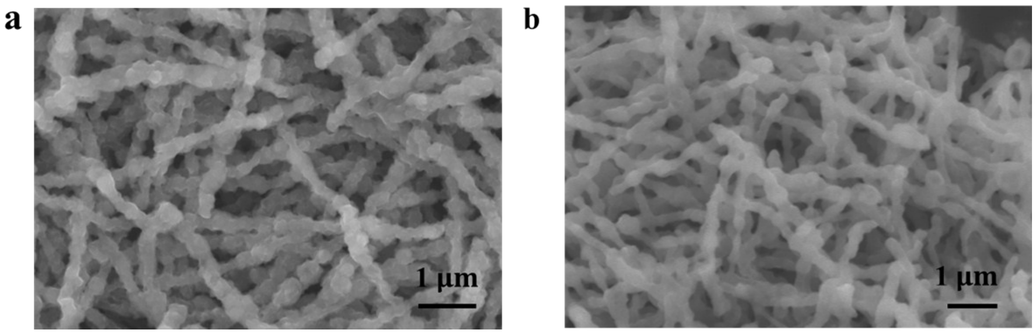

Figure S17. SEM images of the NHC_2-NH_3/Ar electrode after (a) 100 and (b)1000 cycles.

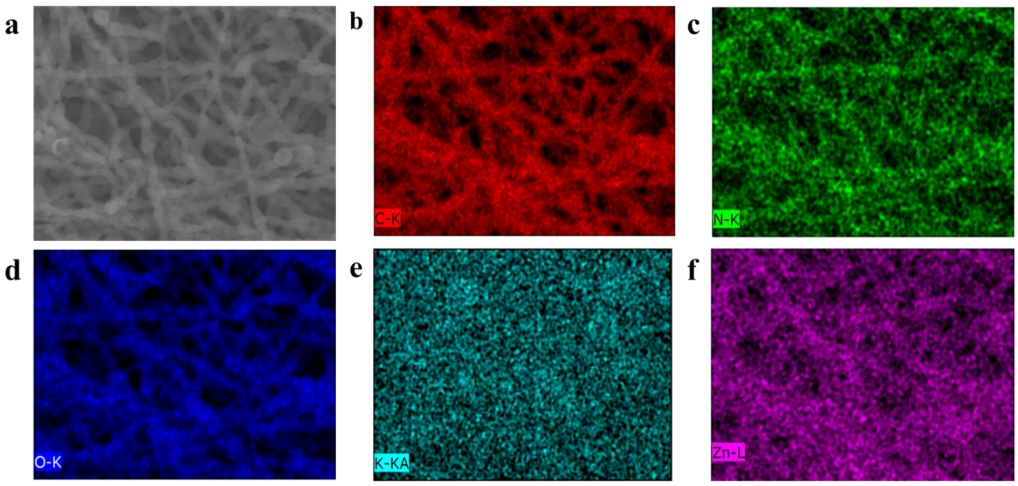

Figure S18. (a) SEM image and (b-f) the EDS mappings of the NHC_2-NH_3/Ar electrode after 100 cycles.

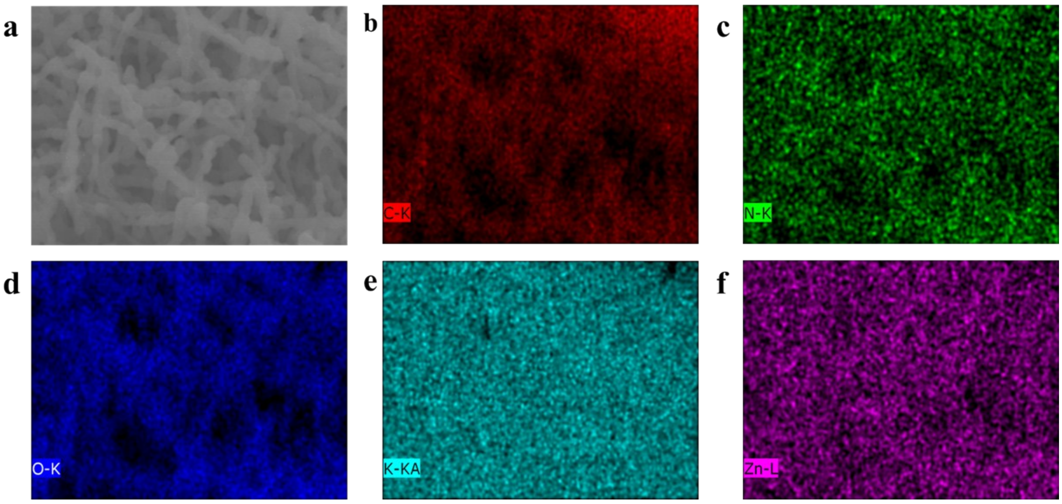

Figure S19. (a) SEM image and (b-f) the EDS mappings of the NHC_2-NH_3/Ar electrode after 1000 cycles.

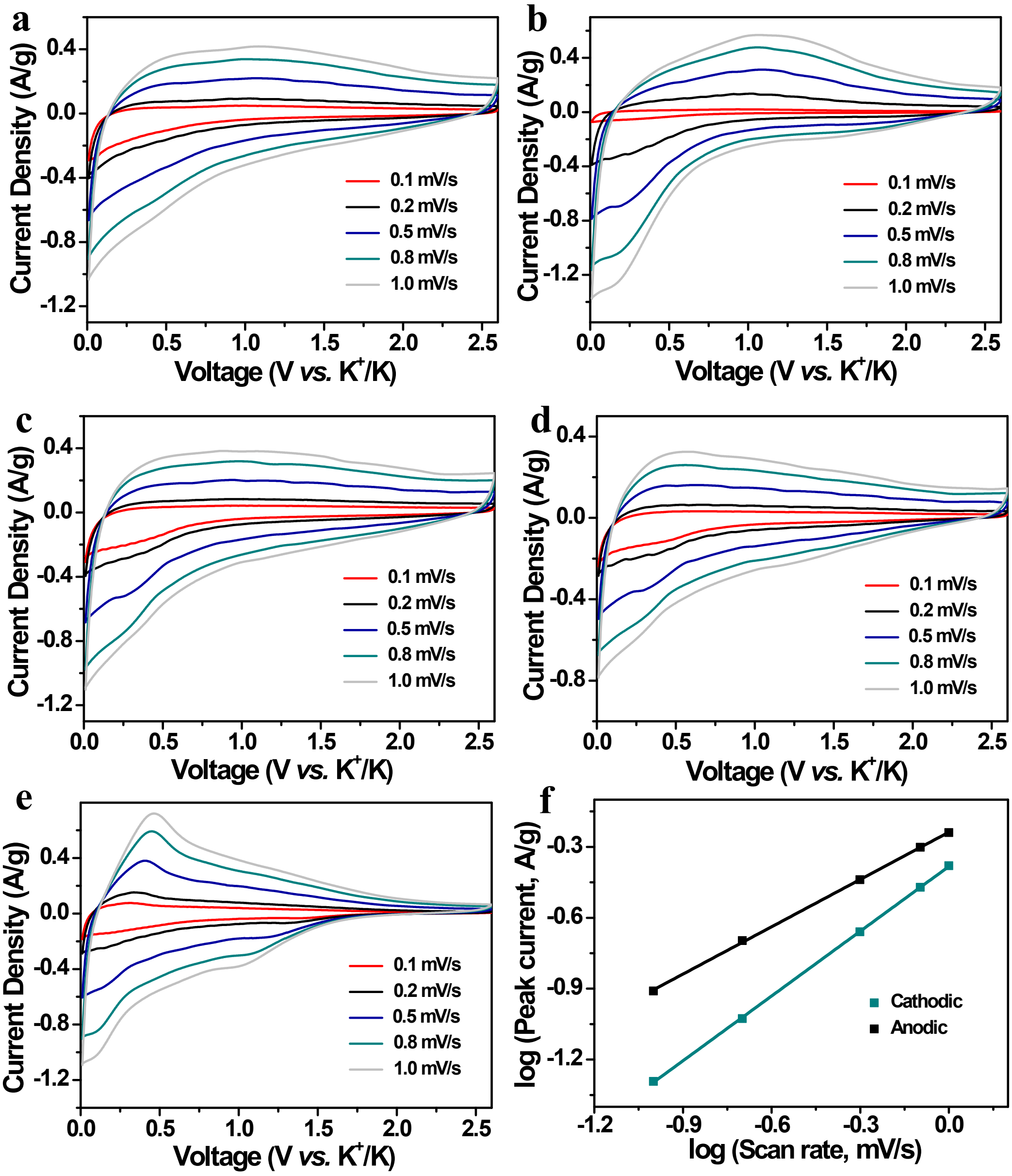

Figure S20 CV profiles of the series of NHCFs materials at different scan rates: (a) NHC_2-NH_3/Ar, (b) NHC_1-NH_3/Ar, (c) NHC_2-Ar-800, (d) NHC_2-Ar-900 and (e) PAN-NH_3/Ar. (f) Determination of the b-value of NHC_2-NH_3/Ar using the relationship between peak current and scan rate in PIBs.

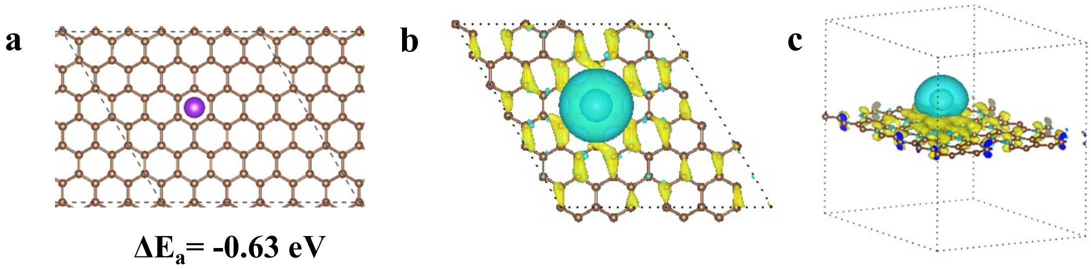

Figure S21 Theoretical simulations and relative verifications. (a) K atom was absorbed in the pristine carbon structure. Side and top views of electron density differences of K absorbed in the pristine carbon structures. Yellow and blue areas represent increased and decreased electron density, respectively. Brown and purple balls represent C and K atoms, respectively

Table S1 Characteristics (XPS and BET surface area) of the different NHC materials.

Materials	C (at. %)	N (at. %)	O (at. %)	N type			BET surface area (m^2 g^{-1})
				N5 (%)	N6 (%)	NQ(%)	
NHC_2-600-Ar	75.77	10.91	10.54	25.1	58.4	16.5	17.1
NHC_2-700-Ar	77.71	8.1	11.35	20.0	56.9	23.1	32.3
NHC_2-800-Ar	89.96	4.9	4.71	24.5	37.4	38.1	358.5
NHC_2-900-Ar	91.74	2.8	5.4	15.1	22.6	62.3	379.7
$NHC_{1/2}$-NH_3/Ar	79.17	10.74	8.24	17.6	56.8	25.6	115.0
NHC_1-NH_3/Ar	78.18	12.35	6.62	16.2	56.0	27.8	118.6
NHC_2-NH_3/Ar	78.76	**10.71**	7.71	22.0	64.0	14.0	355.6

Table S2. Comparison of electrochemical performance of the NHC_2-NH_3/Ar anode with those reported previously for PIBs.

Materials	Reversible Capacity & Rate Capability	Cycle life (capacity (retention proportion @ cycle number@mA/g))	Free-standing Electrode (YES or NO?)	References
NHC_2-NH_3/Ar	**293.5 mAh/g @ 100 mA/g** **224 mAh/g @ 1000 mA/g**	**228.4 mAh/g (82.7 % @ 200 cycles @200 mA/g)** **227.3 mAh/g (82 % @ 500 cycles @200 mA/g)** **225.4 mAh/g (81 % @ 1000 cycles @200 mA/g)** **201.4 mAh/g (83.2 % @200 cycles @500 mA/g)** **192.8 mAh/g (80 % @500 cycles @500 mA/g)** **184.0 mAh/g (77 % @1000 cycles @500 mA/g)** **185.4 mAh/g (83.9 % @ 200 cycles @1000 mA/g)** **172.7 mAh/g (78 % @ 500 cycles @1000 mA/g)** **161.3 mAh/g (73 % @ 1600 cycles @1000 mA/g)**	**YES**	**This work**
RGO	200 mAh/g @ 5 mA/g 50 mAh/g @ 100 mA/g	150 mAh /g (88.2% @ 175 cycles @ 10 mA/g)	YES	[1]
MLG	100 mAh/g @ 200 mA/g 80 mAh/g @ 10000 mA/g	95 mAh /g (95%@ 1000 cycles @ 2000 mA/g)	YES	[2]
HCNTs	232 mAh /g @ 100 mA/g 162 mAh /g @ 1600 mA/g	210 mAh /g (90% @ 500 cycles @ 100 mA/g)	YES	[3]
CNFF	240 mAh /g @ 50 mA/g 164 mAh /g @ 1000 mA/g	158 mAh /g (88% @ 2000 cycles @ 1000 mA/g)	YES	[4]
S-RGO	435 mAh /g @ 50 mA/g 224 mAh /g @ 1 A/g	229 mAh /g (~76% @ 500 cycles @ 1 A/g)	YES	[5]

N-FLG	350 mAh/g @ 50 mA/g 200 mAh/g @ 100 mA/g	210 mAh /g (77.8% @ 100 cycles @ 100 mA/g)	YES	[6]
S/O-PCM	226.6 mAh/g @ 50 mA/g 158 mAh/g @ 1000 mA/g	108.4 mAh/g (69% @ 2000 cycles @ 1000 mA/g)	NO	[7]
HINCA	340 mAh/g @ 28 mA/g ~120 mAh/g @ 560 mA/g	~150 mAh/g (75% @ 500 cycles @ 280 mA/g)	NO	[8]
NOHPHC	365 mAh/g @ 25 mA/g 118 mAh/g @ 3000 mA/ g	230.6 mAh/g (76.1%@ 100 cycles @ 50 mA/g)	NO	[9]
NCNF	238 mAh/g @ 100 mA/g 126 mAh/g @ 5000 mA/g	248 mAh/g (90.8%@100 cycles @ 25 mA/g) 146 mAh/g (100% @ 4000 cycles @ 2 A/g)	NO	[10]
HCS-SC	261 mAh/g @ 27.9 mA/g 190 mAh/g @ 558 mA/g	213.6 mAh/g (89% @ 440 cycles @ 58.8 mA/g) 195.3 mAh/g (93% @ 200 cycles @ 279 mA/g)	NO	[11]
Soft carbon	273 mAh/g @ 6.975 mA/g 140 mAh/g @ 1395 mA/g	150.6 mAh/g (81.4%@ 50 cycles @ 558 mA/g)	NO	[12]
HCM	250 mAh/g @ 0.028 A/g 130 mAh/g @ 1.04 A/g	216 mAh/g(83 % @ 100 cycles @ 0.028 A/g	NO	[13]
amorphous OMC	286.4 mA h/ g @0.05 A/g 144.2 mA h/ g @1.0 A/g	146.5 mAh/g(70% @1000 cycles@1 A/g)	NO	[14]
PNCM	388 mA h/ g @50 mA/g 178 mA h/ g @5000 mA/g	152 mAh/g(67.5% @3000 cycles@1A/g)	NO	[15]
TBMC	500 mA h/g @ 0.05 A/g 136 mA h/g @ 2 A/g	244 mA h /g (83%@ 200 cycles@ 0.5 A/ g)	NO	[16]
HNPC	419.7 mAh/g @ 50 mA/g 185.0 mAh/g @ 10000 mA/g	160.5 mAh/g (81.1% @ 1000 cycles @ 5000 mA/g)	NO	[17]
NPC	587.6 mA h/g @ 50 mA/g 186.2 mA h/g @ 2000 mA/g	231.6 mA h/g (~77%@ 2000 cycles @ 500 mA/ g)	NO	[18]
N-CNS	367 mA h/g @ 50 mA/g	225 mA h/g (~70.5%@ 1000 cycles @ 0.5A/ g)	NO	[19]

	168 mA h/g @ 2000 mA/g	151 mA h/g (~75.6%@ 1000 cycles @ 1 A/ g)		
NBCNTs	359 mA h/g @ 100 mA/g 186 mA h/g @1000 mA/g	204 mA h/g (@ 1000 cycles @ 0.5A/ g)	NO	[20]

Note:

RGO: Reduced graphene oxide

MLG: multi-layered graphene

HCNTs: hierarchical carbon nanotube

CNFF: carbon nanofiber foam

S-RGO: Sulphur-doped reduced graphene oxide

N-FLG: N-doped few-layer graphene

S/O-PCM: sulfur (S) and oxygen (O) codoped porous hard carbon microspheres

HINCA: hollow interconnected neuron-like carbon architecture

NOHPHC: nitrogen/oxygen dual-doped hierarchical porous hard carbon

NCNF: N-doped carbon nanofibers

HCS-SC: hard carbon spheres - soft carbon

HCM: Hard carbon microshperes

OMC: ordered mesoporous carbon

PNCM: pyridinic N-doped porous carbon monolith

TBMC: nanotube-backboned mesoporous carbon

HNPC: hierarchically nitrogen-doped porous carbon

NPC: N-doped porous carbon

N-CNS: Nitrogen doped carbon nanosheets

NBCNTs: nitrogen-doped bamboo-like carbon nanotubes

Reference:

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