

## Supporting information

### **Multi-scale carbon@Sb mesoporous composites activated by in-situ localized electrochemical pulverization as high-rate and long-life anode materials for potassium-ion batteries**

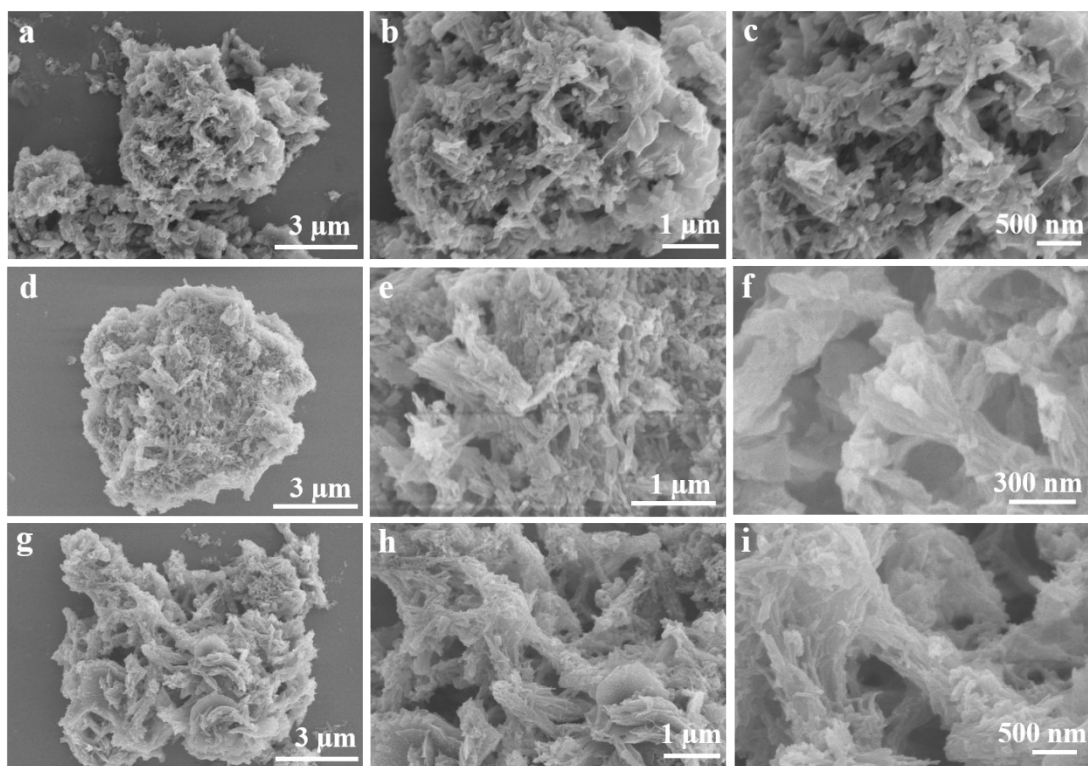
Jie Ren<sup>1</sup>, Xiang Wang<sup>1</sup>, Jihao Li<sup>2</sup>, Qianzi Sun<sup>1</sup>, Shaozhou Li<sup>1</sup>, Ling Bai<sup>1</sup>, Xianming Liu<sup>3</sup>, Guilong Liu<sup>3</sup>, Ziquan Li<sup>1,\*</sup>, Haijiao Zhang<sup>2,\*</sup>, Zhen-Dong Huang<sup>1,\*</sup>

1. State Key Laboratory for Organic Electronics and Information Displays & Jiangsu Key Laboratory for Biosensors, Institute of Advanced Materials, Jiangsu National Synergetic Innovation Center for Advanced Materials, Nanjing 210023, P. R. China

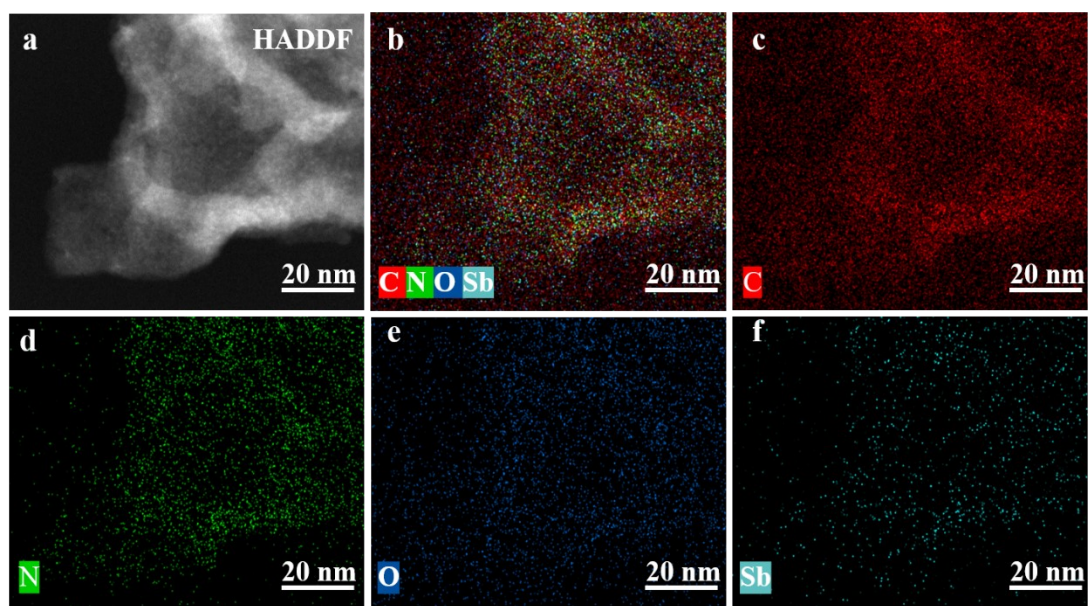
2. Institute of Nanochemistry and Nanobiology, School of Environmental and Chemical Engineering, Shanghai University, Shanghai 200444, P. R. China

3. College of Chemistry and Chemical Engineering, Luoyang Normal University, Luoyang, Henan, 471934, P. R. China

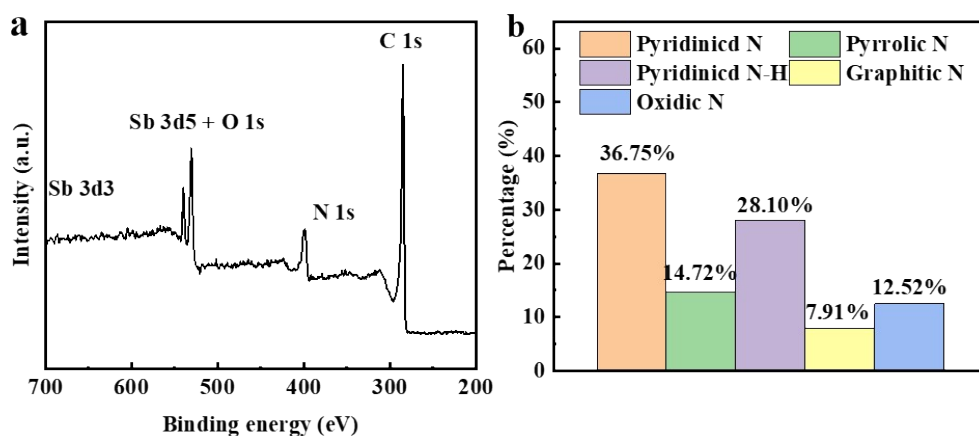
\*Corresponding authors: [iamzdhuang@njupt.edu.cn](mailto:iamzdhuang@njupt.edu.cn) (Z. D. Huang); [lizq@njupt.edu.cn](mailto:lizq@njupt.edu.cn) (Z. Q. Li); [hjzhang128@shu.edu.cn](mailto:hjzhang128@shu.edu.cn) (H. J. Zhang)



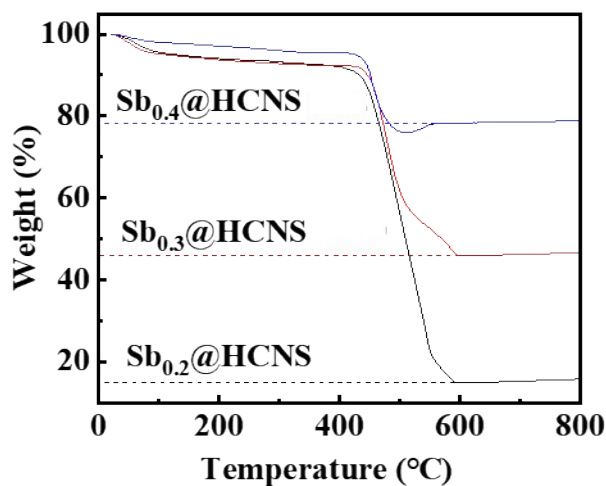
**Figure S1.** SEM images in different magnifications of samples: (a-c)  $\text{Sb}_2@\text{DCDA}/\text{PACP}$ , (d-f)  $\text{Sb}_3@\text{DCDA}/\text{PACP}$ , and (g-i)  $\text{Sb}_4@\text{DCDA}/\text{PACP}$ .



**Figure S2.** (a) HAADF image of  $\text{Sb}_3@\text{HCNS}$ , and (b-f) combined and individual elemental distribution maps of C, N, O, and Sb.

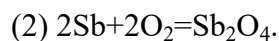
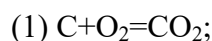


**Figure S3.** (a) X-ray photoelectron spectroscopy (XPS) full spectrum of  $\text{Sb}_3\text{@HCNS}$ , (b) The fraction of pyridinic N (398.2 eV), pyrrolic N (399.6 eV), pyridinic N-H (400.7 eV), graphitic N (401.5 eV), and oxidized N (403.5 eV) within N 1s spectrum in Figure 3f.



**Figure S4.** Thermogravimetry (TG) curves of as-prepared Sb<sub>2</sub>@HCNS, Sb<sub>3</sub>@HCNS, and Sb<sub>4</sub>@HCNS.

The weight loss of carbon combustion and the weight gain of Sb<sub>2</sub>O<sub>4</sub> formation based on the following reactions:



The weight ratio of Sb in the nanocomposite can be calculated based on the following equation:

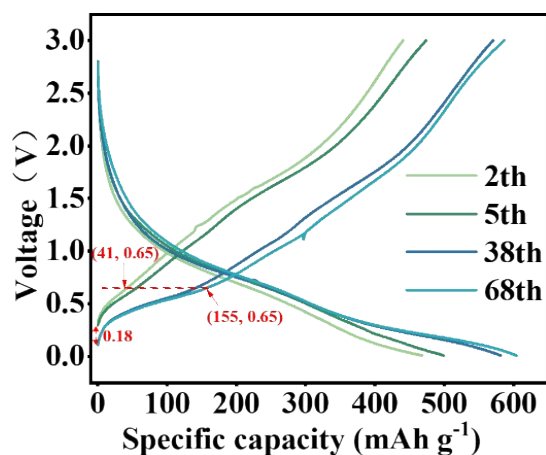
$$Sb(wt. \%) = \frac{\text{molecular weight of Sb}}{\text{molecular weight of Sb}_2\text{O}_4} \times \frac{\text{final weight of Sb}_2\text{O}_4}{\text{initial weight of Sb@HCNS}}$$

Equation S1

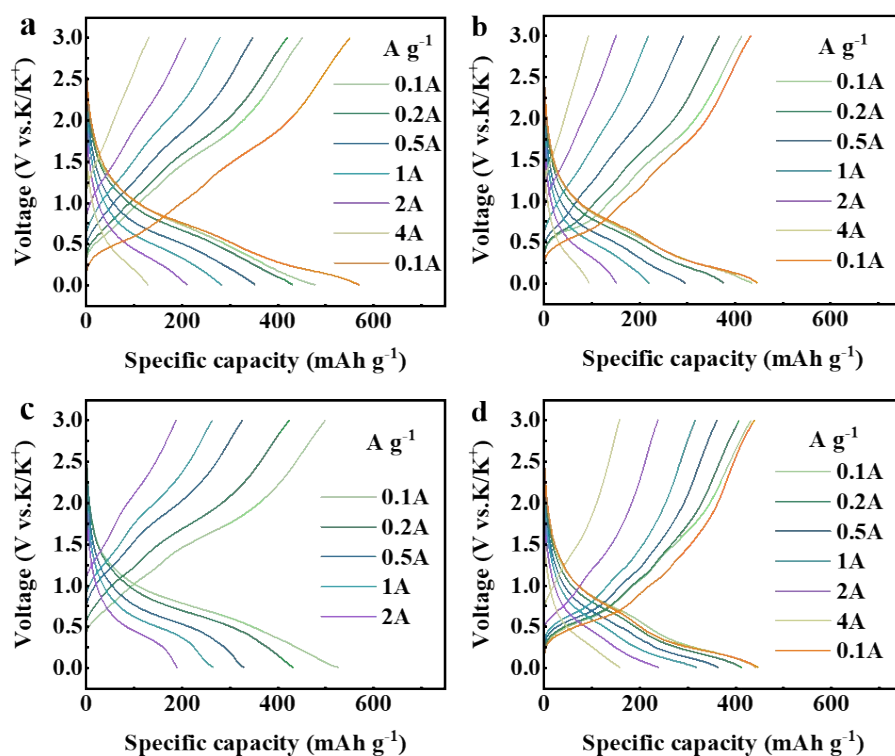
$$Sb_2@HCNS: S(wt. \%) = \frac{121.76}{307.52} \times \frac{15.1}{100} \times 2 \times 100\% = 11.96\%$$

$$Sb_3@HCNS: S(wt. \%) = \frac{121.76}{307.52} \times \frac{45.9}{100} \times 2 \times 100\% = 36.35\%$$

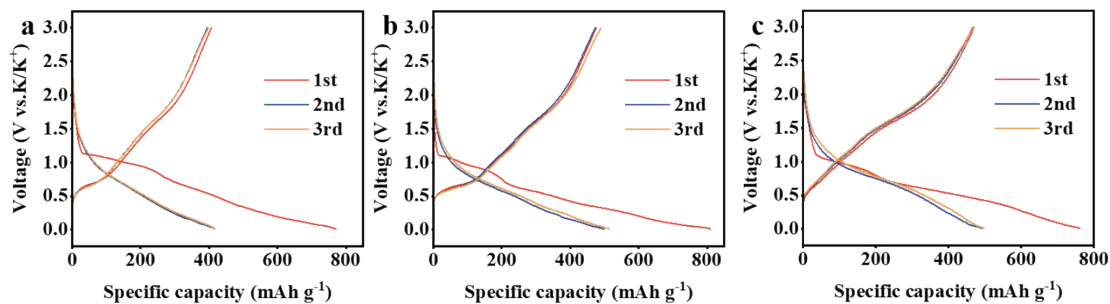
$$Sb_4@HCNS: S(wt. \%) = \frac{121.76}{307.52} \times \frac{78.5}{100} \times 2 \times 100\% = 62.16\%$$



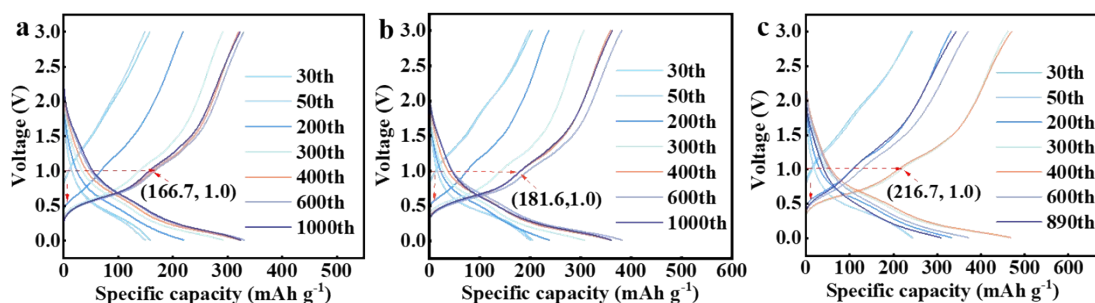
**Figure S5.** The discharge/charge profiles of  $\text{Sb}_3\text{@HCNS}$  at the 2nd, the 5th, the 38th and the 68th cycle at the current density of  $0.1 \text{ A g}^{-1}$  during the rate capability testing.



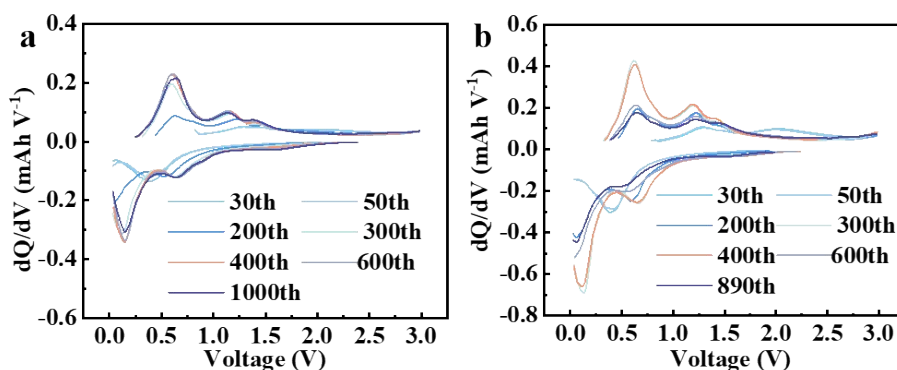
**Figure S6.** The typical charge/discharge profiles of (a)  $\text{Sb}_2\text{@HCNS}$ , (b)  $\text{Sb}_3\text{@HCNS}$ , (c)  $\text{Sb}_4\text{@HCNS}$  at various current densities at 2nd, 7th, 12th, 17th, 22nd, 27th and 32nd, respectively. (d) The corresponding discharge/charge profiles of  $\text{Sb}_2\text{@HCNS}$  at 38th, 43rd, 48th, 53rd, 58th, and 63rd.



**Figure S7.** The discharge/charge profiles of  $\text{Sb}_2@\text{HCNS}$  (a),  $\text{Sb}_3@\text{HCNS}$  (b), and  $\text{Sb}_4@\text{HCNS}$  (c) at the current density of  $0.1 \text{ A g}^{-1}$  during the initial three activation cycles before long-term cyclic testing at  $2 \text{ A g}^{-1}$ .

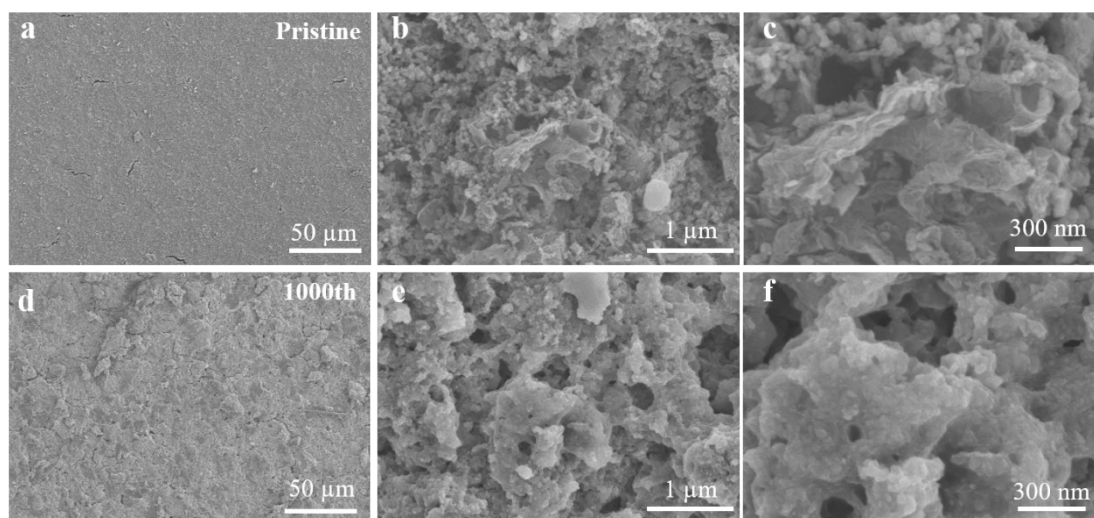


**Figure S8.** The different cycle discharge/charge profiles of  $\text{Sb}_2@\text{HCNS}$  (a),  $\text{Sb}_3@\text{HCNS}$  (b), and  $\text{Sb}_4@\text{HCNS}$  (c) at the current density of  $2 \text{ A g}^{-1}$  during the long-term cyclic testing.

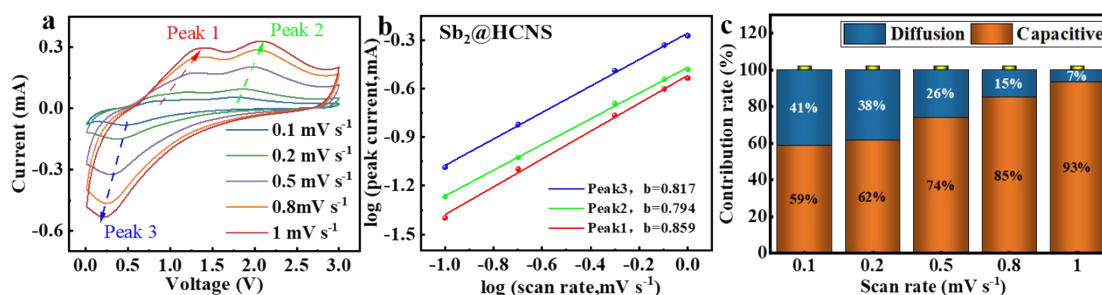


**Figure S9.** The  $dQ/dV$  profiles of  $\text{Sb}_2@\text{HCNS}$  (a) and  $\text{Sb}_4@\text{HCNS}$  (b) corresponding to the different cycle discharge/charge profiles at the current density of  $2 \text{ A g}^{-1}$  during the long-term cyclic testing shown in Figures S7a and S7c.





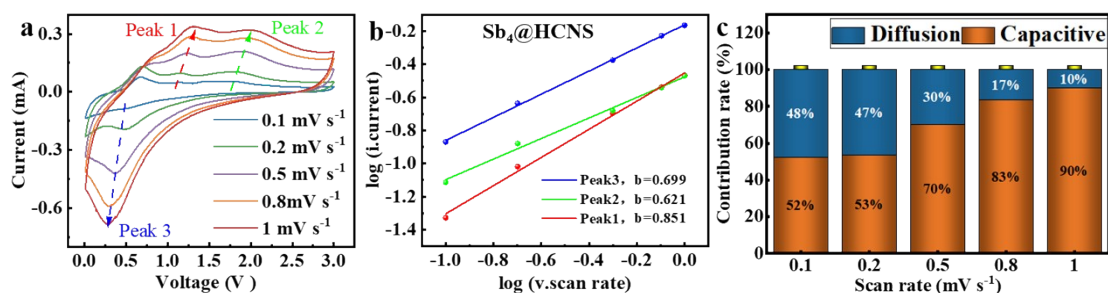
**Figure S10.** SEM images of (a-c) pristine  $\text{Sb}_3@\text{HCNS}$  electrode, and (d-f) the  $\text{Sb}_3@\text{HCNS}$  electrode being cycled for 1000 cycles at  $2 \text{ A g}^{-1}$ .



**Figure S11.** (a) CV curves of  $\text{Sb}_2@\text{HCNS}$  at various scan rates from 0.1 to 1.0  $\text{mV s}^{-1}$ . (b) The plot of  $\log(\text{peak current})$  versus  $\log(\text{scan rate})$  of redox peaks from CV curves and b-value determination lines. (c) Contribution ratios of the surface capacitance and inside diffusion-based capacity to the total capacity of  $\text{Sb}_2@\text{HCNS}$  at different scan rates.

The contributions of the surface-controlled and diffusion-controlled reactions were quantitatively estimated based on the redox peak currents of CV curves according to

the equation:  $i = av^b$ . Normally, the peak current,  $i$ , is proportional to the sweep rate,  $v$ . The  $b$  value was determined from the plot slope of the  $\log(i)$  versus  $\log(v)$ . When the  $b$  value is 1, the electrochemical reaction is purely surface-controlled. when the  $b$  value is 0.5, the electrochemical reaction is entirely diffusion-controlled. The surface-controlled ( $k_1v$ ) and diffusion-controlled ( $k_2v^{1/2}$ ) contribution to final capacity can be divided into components according to the equation:  $i = k_1v + k_2v^{1/2}$  [1].



**Figure S12.** (a) CV curves of Sb<sub>4</sub>@HCNS at various scan rates from 0.1 to 1.0 mV s<sup>-1</sup>. (b) The plot of log (peak current) versus log (scan rate) of redox peaks from CV curves and b-value determination lines. (c) Contribution ratios of the surface capacitance and inside diffusion-based capacity to the total capacity of Sb<sub>4</sub>@HCNS at different scan rates.



**Table S1.** HRTEM EDX analysis of C, N, O, Sb atomic and mass fraction in Sb<sub>3</sub>@HCNS.

Element	Atomic fraction (%)	Mass fraction (%)
C	83.83	73.39
N	10.15	10.36
O	4.82	5.62
Sb	1.20	10.63

**Table S2.** XPS analysis of C, N, O, Sb atomic and mass fraction in Sb<sub>3</sub>@HCNS.

Element	Atomic fraction (%)	Mass fraction (%)
C	71.15	56.26
N	13.73	12.67
O	12.95	13.65
Sb	2.17	17.42%

**Table S3.** The performance comparison of Sb<sub>3</sub>@HCNS with the reported counterparts.

Materials	Cyclability (mAh g <sup>-1</sup> )	Rate performance (mAh g <sup>-1</sup> )	Ref.
Sb SQ@MA	~314 at 1 A g <sup>-1</sup> after 1000 cycles	~447 at 0.2 A g <sup>-1</sup>	[37]
		~419 at 0.4 A g <sup>-1</sup>	
		~366.4 at 0.8 A g <sup>-1</sup>	
		~299 at 1.6 A g <sup>-1</sup>	
		~246 at 3.2 A g <sup>-1</sup>	
		~379 at 0.1 A g <sup>-1</sup>	
Sb-N-C	~254 at 1 A g <sup>-1</sup> after 2000 cycles	~343 at 0.2 A g <sup>-1</sup>	[39]
		~302 at 0.5 A g <sup>-1</sup>	
		~274 at 1 A g <sup>-1</sup>	
		~245 at 2 A g <sup>-1</sup>	
O-Sb-N SA@NC2	~195 at 2A g <sup>-1</sup> after 2000 cycles	~576 at 0.1 A g <sup>-1</sup>	[41]
		~536 at 0.2 A g <sup>-1</sup>	
		~312 at 0.5 A g <sup>-1</sup>	
		~238 at 1 A g <sup>-1</sup>	
		~225 at 2 A g <sup>-1</sup>	
		~166 at 4 A g <sup>-1</sup>	
Sb SA/C	~331.3 at 1A g <sup>-1</sup> after 1100 cycles	~441 at 0.1 A g <sup>-1</sup>	[48]
		~418 at 0.2 A g <sup>-1</sup>	
		~356 at 0.5 A g <sup>-1</sup>	
		~312 at 1 A g <sup>-1</sup>	
		~252 at 2 A g <sup>-1</sup>	
NPS-FCM	~162.1 at 1 A g <sup>-1</sup> after 1000cycles	~231.2 at 0.1 A g <sup>-1</sup>	[49]
		~205.7 at 0.3 A g <sup>-1</sup>	
		~190.4 at 0.5 A g <sup>-1</sup>	
		~179.4 at 1 A g <sup>-1</sup>	
HTSb@Sb <sub>2</sub> O <sub>3</sub> @C	~241.67 at 2 A g <sup>-1</sup> after 2000 cycles	~162.7 at 2 A g <sup>-1</sup>	[50]
		~442.9 at 0.1 A g <sup>-1</sup>	
		~420.2 at 0.2 A g <sup>-1</sup>	
		~361.4 at 0.5 A g <sup>-1</sup>	
		~311.3 at 1 A g <sup>-1</sup>	
		~273 at 2 A g <sup>-1</sup>	

---

		~284 at 0.1 A g <sup>-1</sup>	
		~258 at 0.2 A g <sup>-1</sup>	
Mn-NC	~146 at 1 A g <sup>-1</sup> after 3000cycles	~240 at 0.4 A g <sup>-1</sup>	[51]
		~207 at 0.8 A g <sup>-1</sup>	
		~191 at 1 A g <sup>-1</sup>	
		~162 at 2 A g <sup>-1</sup>	
		~417 at 0.1 A g <sup>-1</sup>	
		~329 at 0.2 A g <sup>-1</sup>	
Ni @BNHC	~279 at 1A g-1 after 1500cycles	~268 at 0.5 A g <sup>-1</sup>	[52]
		~215 at 1 A g <sup>-1</sup>	
		~154 at 2 A g <sup>-1</sup>	
		~230 at 0.1 A g <sup>-1</sup>	
		~201 at 0.2 A g <sup>-1</sup>	
SCNs	~80 at 5 A g <sup>-1</sup> after 5000cycles	~183 at 0.5 A g <sup>-1</sup>	[53]
		~167 at 1 A g <sup>-1</sup>	
		~126 at 2 A g <sup>-1</sup>	
		~604 at 0.1 A g <sup>-1</sup>	
		~550 at 0.2 A g <sup>-1</sup>	
Sb@Cu <sub>15</sub> Si <sub>4</sub>	~250.2 at 0.2 A g <sup>-1</sup> after	~495 at 0.5 A g <sup>-1</sup>	[54]
NW	1250cycles	~321 at 1 A g <sup>-1</sup>	
		~228.4 at 2 A g <sup>-1</sup>	
		~104.9 at 4 A g <sup>-1</sup>	
		~274.6 at 0.05A g-1	
		~264.2 at 0.1A g-1	
NS-SGNR	~224 at 0.5A g-1 after 500cycles	~257.5 at 0.2A g-1	[55]
		~248.3 at 0.5A g-1	
		~237.4 at 1A g-1	
		~211.7 at 2A g-1	
		~239 at 0.1 A g <sup>-1</sup>	
		~200 at 0.2 A g <sup>-1</sup>	
QLGC	~200at 0.1 A g <sup>-1</sup> after 100cycles	~108at 0.5 A g <sup>-1</sup>	[56]
		~63 at 1 A g <sup>-1</sup>	
		~28.4 at 2 A g <sup>-1</sup>	
		~500 at 0.1 A g <sup>-1</sup>	

---

		~430 at 0.2 A g <sup>-1</sup>	
		~373 at 0.3 A g <sup>-1</sup>	
NP-Sb	~318 at 0.1 A g <sup>-1</sup> after 50cycles	~312 at 0.4 A g <sup>-1</sup>	[57]
		~265 at 0.5 A g <sup>-1</sup>	
		~362 at 0.1 A g <sup>-1</sup>	
		~314 at 0.2 A g <sup>-1</sup>	
Defect-rich C	~321 at 0.05 A g <sup>-1</sup> after 400cycles	~267 at 0.5 A g <sup>-1</sup>	[58]
		~240 at 1 A g <sup>-1</sup>	
		~215 at 1 A g <sup>-1</sup>	
		~437 at 0.1 A g <sup>-1</sup>	
		~400 at 0.2 A g <sup>-1</sup>	
Sb <sub>2</sub> O <sub>3</sub> @PCN	~3437 at 0.1 A g <sup>-1</sup> after 50cycles	~337 at 0.5 A g <sup>-1</sup>	[59]
		~210 at 1 A g <sup>-1</sup>	
		~501 at 0.1 A g <sup>-1</sup>	
		~480 at 0.2 A g <sup>-1</sup>	
Sb@CTF-NC	~440 at 0.05 A g <sup>-1</sup> after 2000cycles	~402 at 0.5 A g <sup>-1</sup>	
		~351 at 1 A g <sup>-1</sup>	[60]
		~232 at 2 A g <sup>-1</sup>	
		~580.8 at 0.1 A g <sup>-1</sup>	
		~540.6 at 0.2 A g <sup>-1</sup>	
Sb <sub>3</sub> @HCNS	~382 at 2 A g <sup>-1</sup> after 1000 cycles	~477.5 at 0.5 A g <sup>-1</sup>	The
		~413.0 at 1 A g <sup>-1</sup>	work
		~325 at 2 A g <sup>-1</sup>	
		~215.5 at 4 A g <sup>-1</sup>	