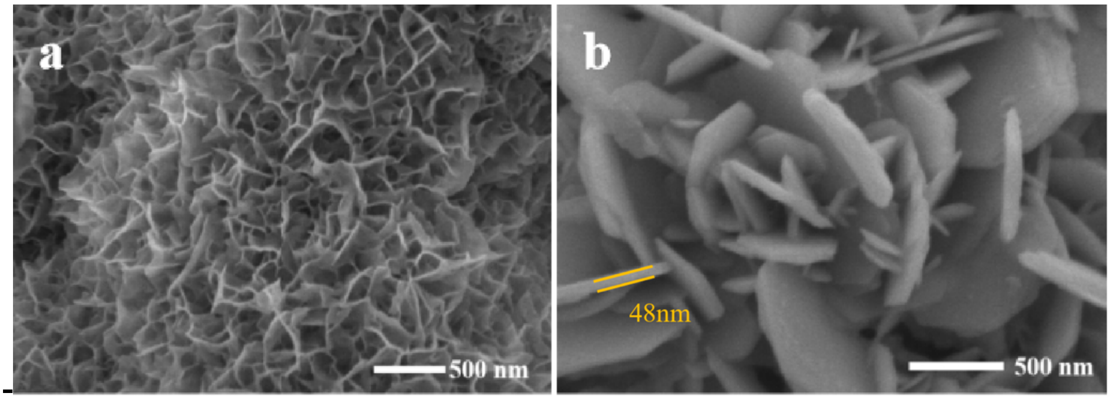

Fig. S1. SEM images of the (a) $Ni(OH)_2$ nanosheets and (b) $NiPS_3$ nanasheets.

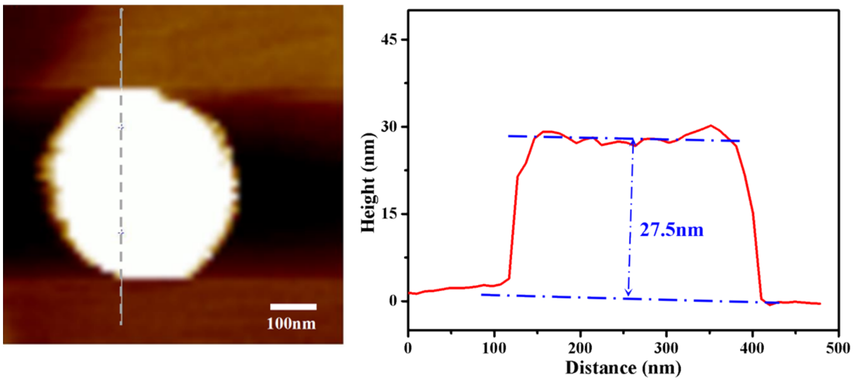

Fig. S2. AFM image of $NiPS_3$ nanosheets.

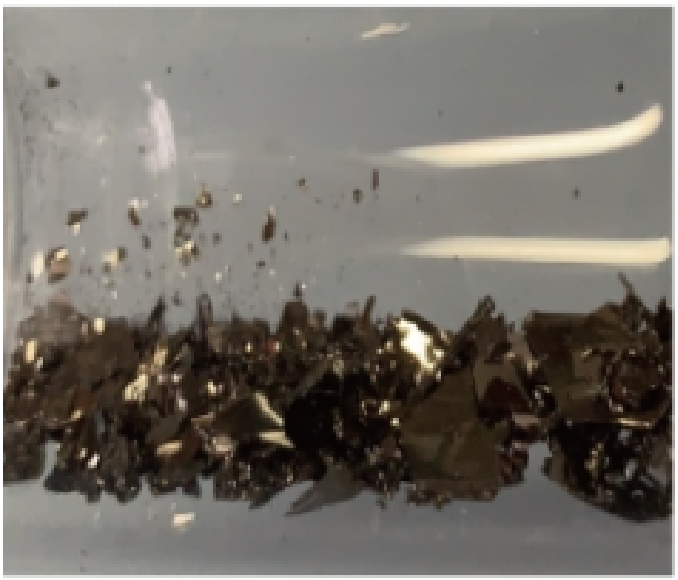

Fig. S3. Image of the bulk $NiPS_3$ synthesized by traditional method. Stoichiometric ratios of Ni, P and S were placed in vacuum ampoules and heated in a dual temperature zone (650°- 700°) tube furnace for five days.

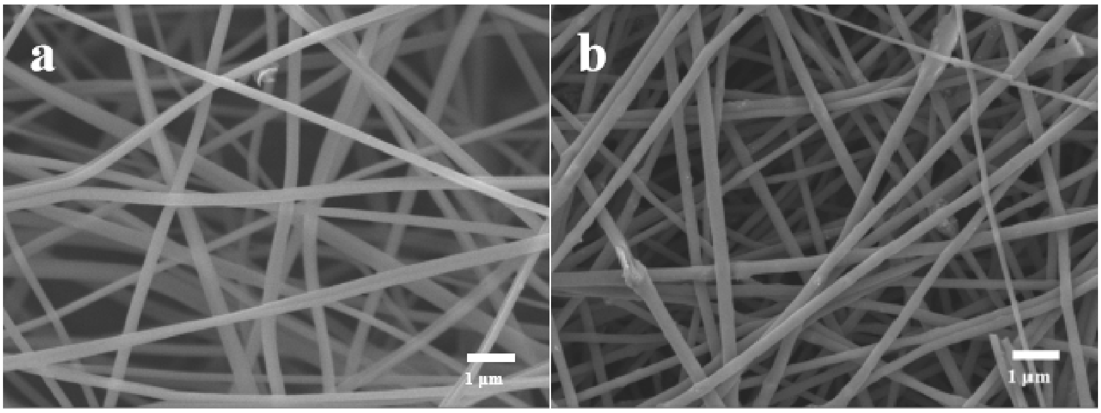

Fig. S4. SEM images of the (a) $NiPS_3$/CNFs and (b) NCNFs.

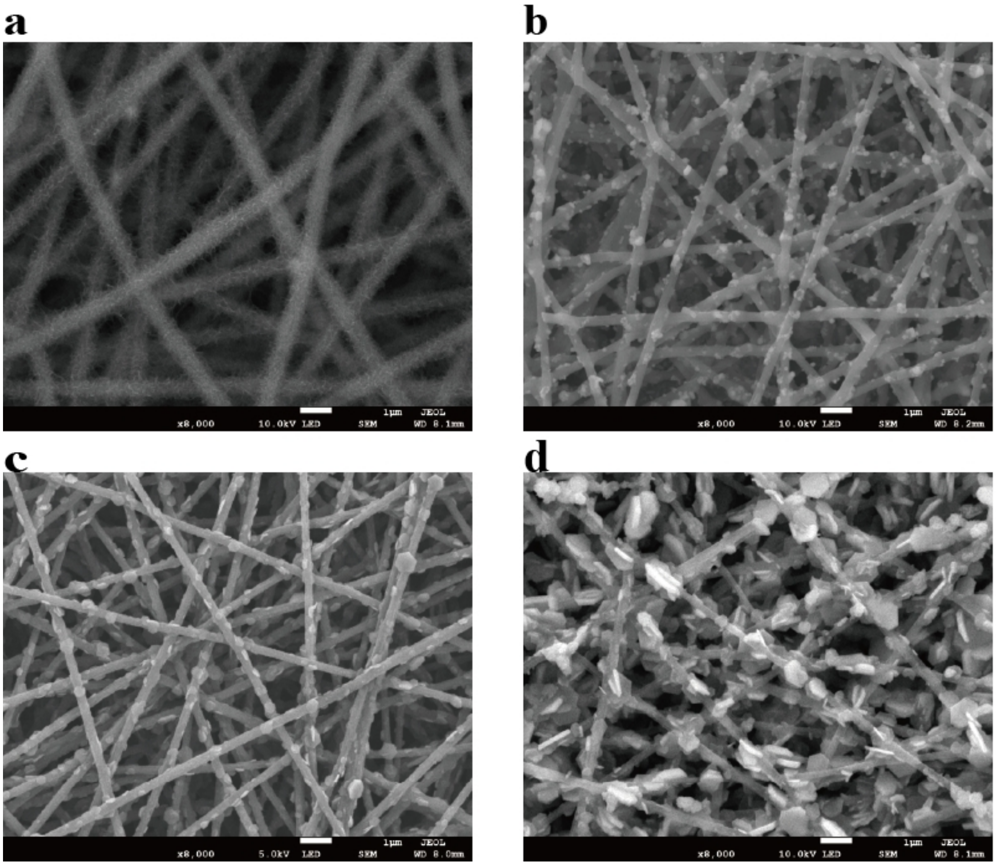

Fig. S5. SEM images of the a) $Ni(OH)_2$@NCNFs; b) $NiPS_3$@NCNFs-1 (0.5mmol $NiSO_4·6H_2O$) , c) $NiPS_3$@NCNFs-2 (1mmol $NiSO_4·6H_2O$) and d) $NiPS_3$@NCNFs-3 (1.5mmol $NiSO_4·6H_2O$). The content of $NiPS_3$ nanosheets were controlled by the concentration of nickel ions.

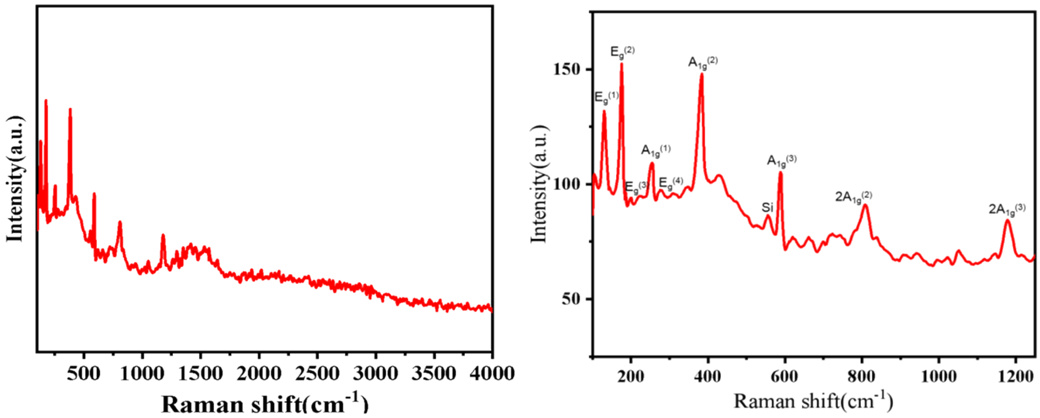

Fig. S6. Raman spectrum of $NiPS_3$@NCNFs, the Raman light wave number is 532nm.

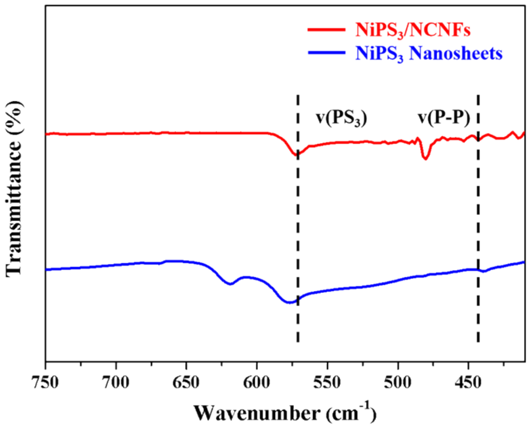

Fig. S7.FT-IR spectrum of $NiPS_3$@NCNFs.

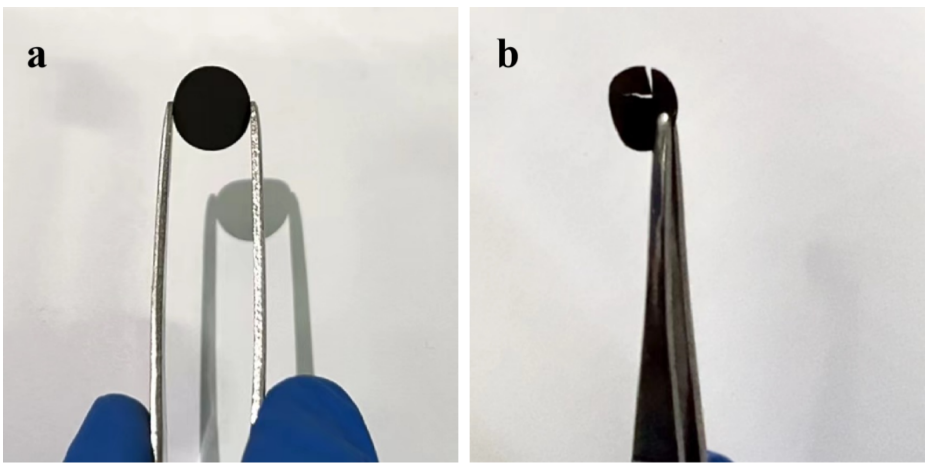

Fig. S8. (a) The $NiPS_3$@NCNFs after being folded in half for 300 times, (b) the cracked $NiPS_3$/CNFs after several folds.

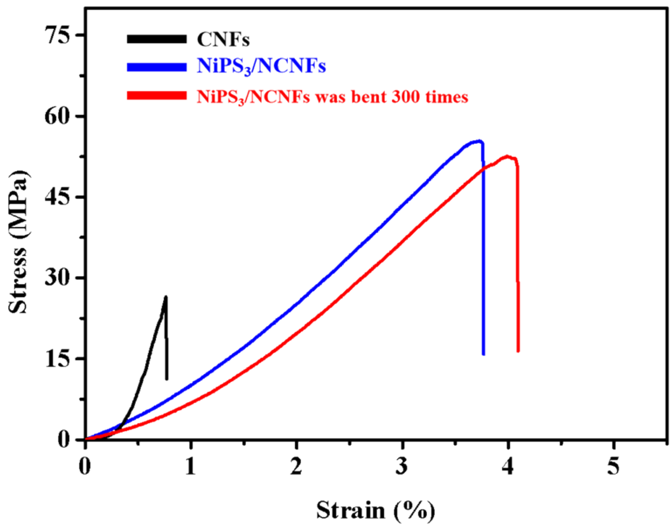

Fig. S9. The stress-strain curves of $NiPS_3$/NCNFs, NCNFs and CNFs.

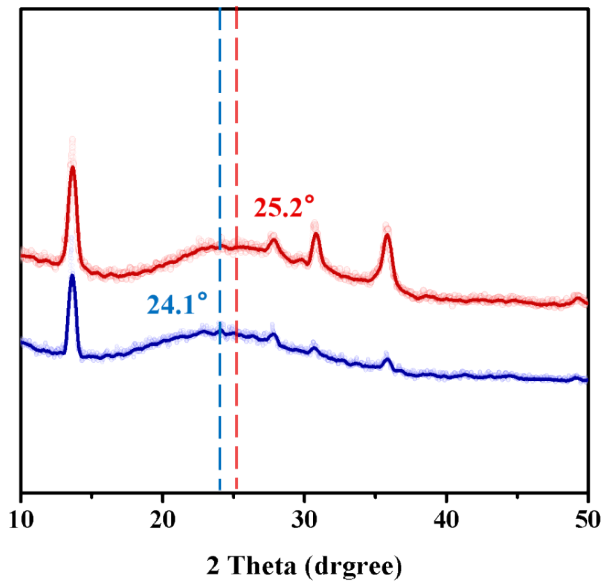

Fig. S10. XRD patterns of $NiPS_3$@CNFs and $NiPS_3$/NCNFs.

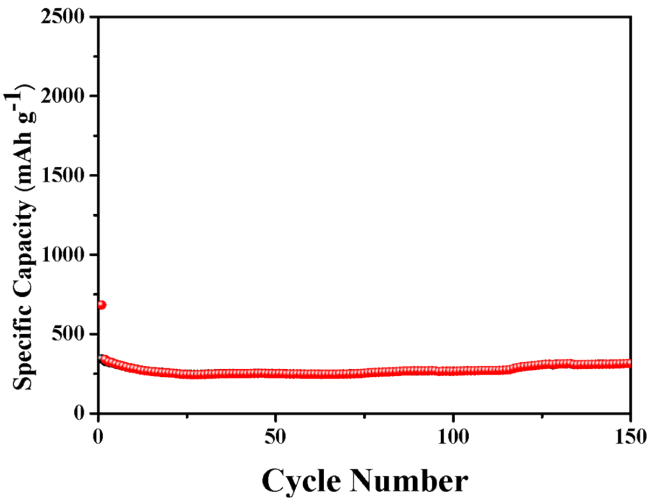

Fig. S11. Cycling performance of the NCNFs electrode at 0.1A g^{-1}.

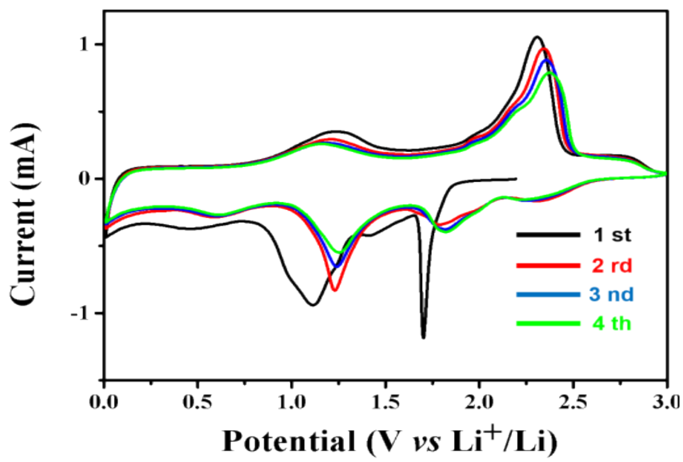

Fig. S12. CV curves of the $NiPS_3$@NCNFs between 0.01 -3 V at a scan rate 0.1 mV/s.

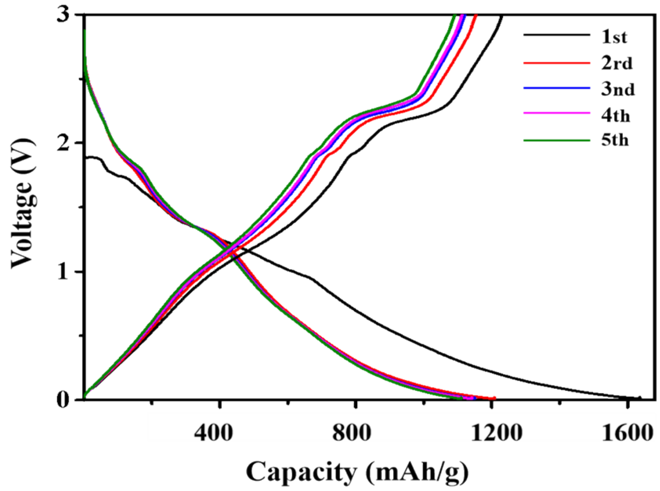

Fig. S13. Galvanostatic charge-discharge curves of the $NiPS_3$@NCNFs.

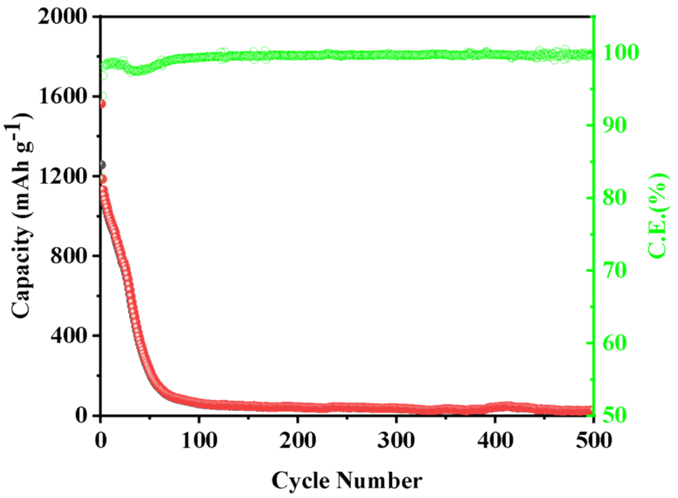

Fig. S14. Cycling performance of bulk $NiPS_3$ electrode at 1A g^{-1}.

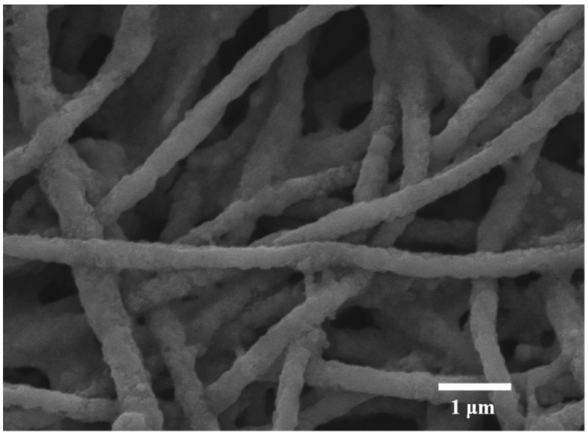

Fig. S15. SEM images of the $NiPS_3$/NCNFs after 1000 times charge-discharge.

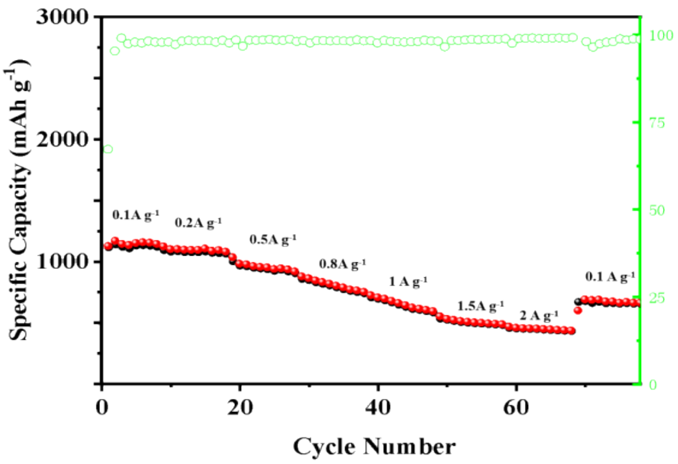

Fig. S16. Rate performances of the $NiPS_3$@NCNFs.

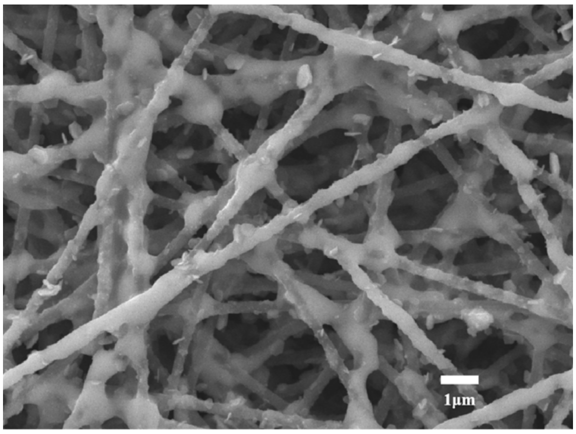

Fig. S17. SEM images of the $NiPS_3$@NCNFs after 3 times charge-discharge.

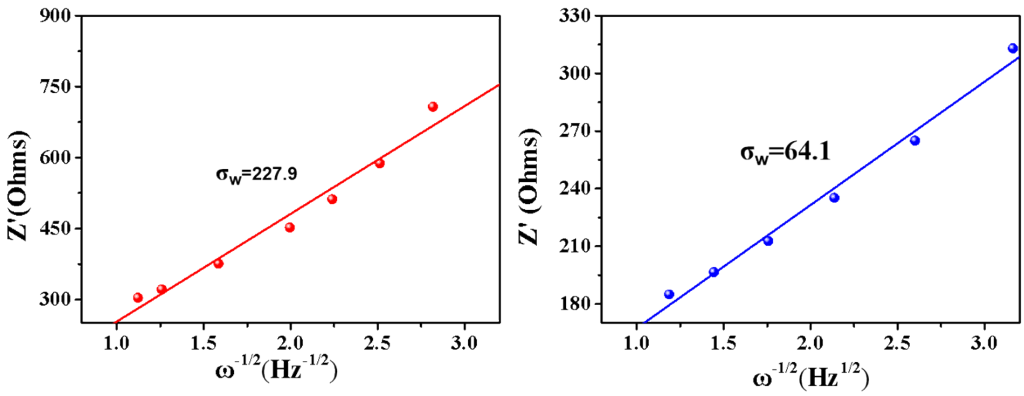

Fig. S18. The plot of real part of the impedance (Z') against the inverse square root of the angular momentum.

Table.S1. The compositions of the samples were determined by ICP. 1 mg sample was first dissolved in hot concentrated nitric acid, and the solution was diluted to 100 mL. A standard solution with Ni^{2+} concentration of 0.5, 1, 1.5, 2, and 2.5 ppm were prepared to get the standard curve.

Samples	Nickel ion concentration	Nickel ion content (per mg in electrode)	$NiPS_3$ content (wt%)
$NiPS_3$@NCNFs-1	0.806mg/L	0.0806mg	25.52%
$NiPS_3$@NCNFs-2	1.088mg/L	0.1088mg	34.45%
$NiPS_3$@NCNFs-3	2.147mg/L	0.2147mg	67.99%
$NiPS_3$/CNFs	1.654mg/L	0.1654mg	52.37%
$NiPS_3$/NCNFs	1.248mg/L	0.1248mg	39.52%

Table.S2. Comparison of $NiPS_3$/NCNFs with other reported freestanding carbon-based electrodes.

Material	Morphology	Flexibility	Specific capacity (mAh g^{-1}) /Current density (A g^{-1})	Retention capacity (mAh g^{-1})/ Cycling number /Current density (A g^{-1})
$NiPS_3$/NCNFs (This work)	$NiPS_3$ nanosheets embedded in NCNF	Folded 300 times without creases	2156.2/0.1	893.5/1000/1
SnO_2/CNF[1]	SnO_2 nanoparticle embedded in CNF	Demonstrate flexible by image	1293/0.1	754/300/1
Porous CNF[2]	Carbon nanofibers	Could be bent without breaking	405/0.03	380/200/0.03
MoS_2/CNF[3]	MoS_2 nanosheets on CNF	Could be bent without breaking	1200/0.1	670/200/0.5
NiS/CNF[4]	NiS nanoparticles on CNF	Demonstrate flexibility by images	1769/0.1	1021/100/0.1
SC-NF[5]	core−shell Si/C nanofiber	Demonstrate flexibility by images	1638/0.1	720/200/0.5
SnS/C[6]	SnS nanoparticles on CNF	Demonstrate flexibility by images	1278/0.1	330/1000/0.8
FeP@C[7]	FeP nanotubes on CNF	Could be bent without breaking	1350/0.1	500/1100/1.5
MnOQD/CNT[8]	MnO quantum dots on CNT	Demonstrate flexibility by images	1361/0.1	883/1000/1
SHCM/NCF[9]	Si nanodots dispersed in carbon	Bent 180° without breaking	2583/0.1	1442/800/1
$NiCoPS_3$/NC[10]	Cubic-like structure	Brittle	1312/0.1	831/1200/2

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