

## Supporting Information

### **Bioinspired Pomegranate-like Microflowers Confining Core-shell Binary $\text{Ni}_x\text{S}_y$ Nanobeads for Efficient Supercapacitors Exhibiting Durable Lifespan Exceeding 100 000 circles**

*Cheng Yang, Minjie Shi, Yuyu Tian, Jun Yang, Xuefeng Song, \* Liping Zhao, Jing Liu, Peng Zhang, Lian Gao\**

C. Yang, Y. Y. Tian, Prof. X. F. Song, Prof. L. P. Zhao, Prof. J. Liu, Prof. P. Zhang, Prof. L. Gao

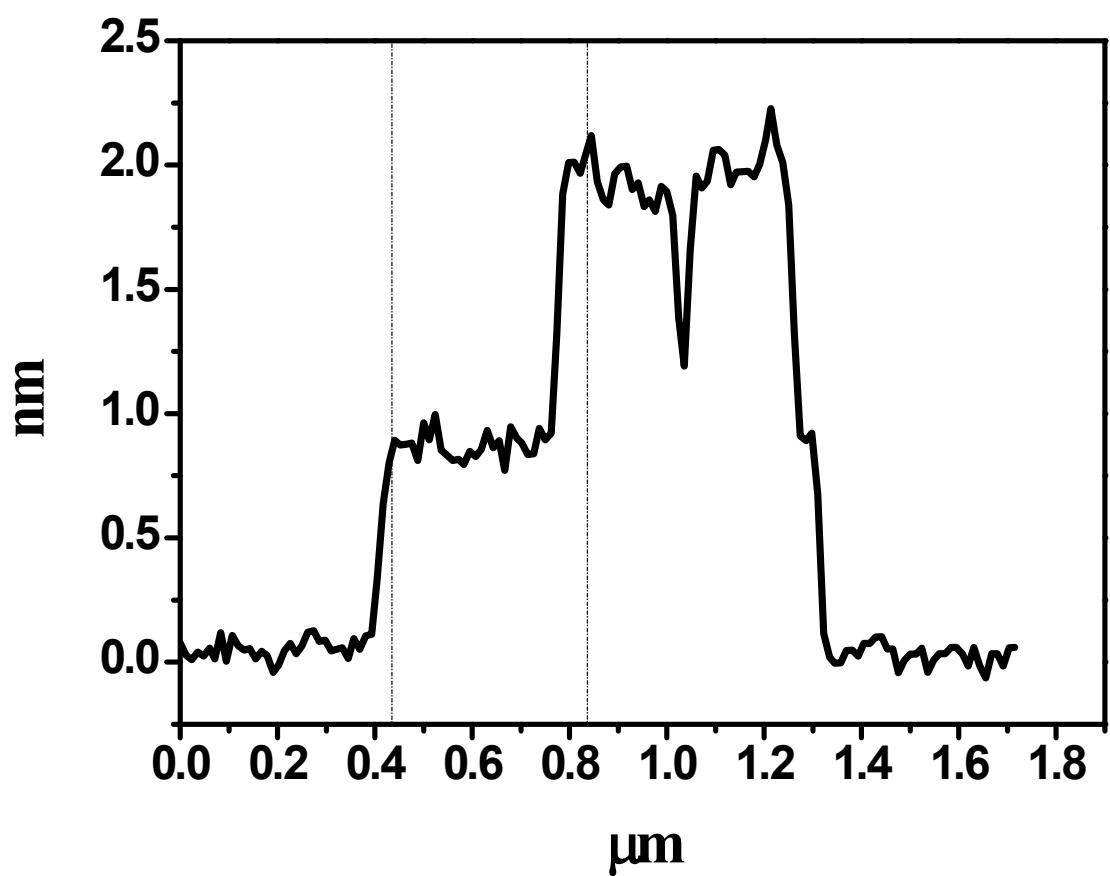
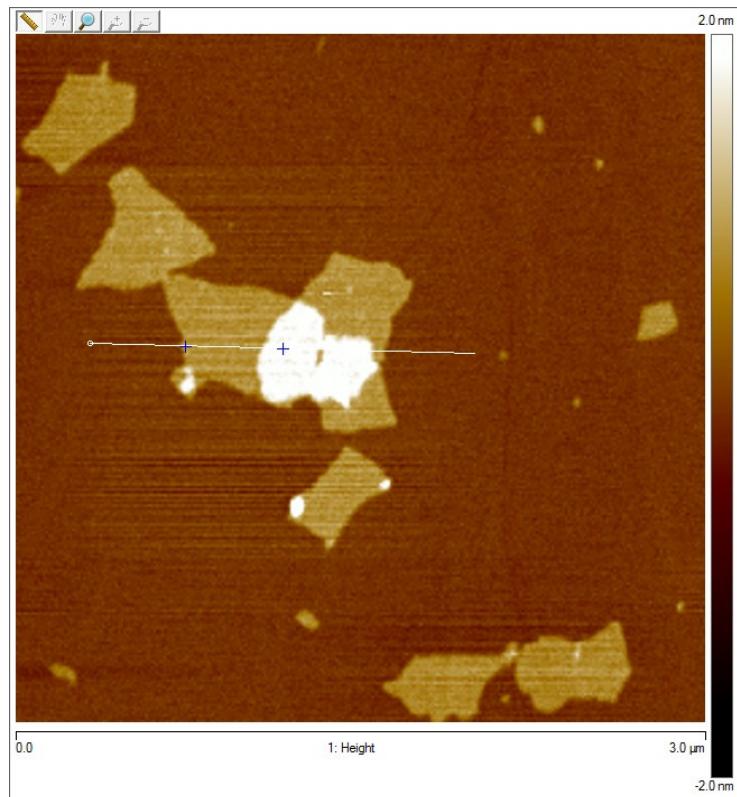
State Key Laboratory for Metallic Matrix Composite Materials, School of Materials Science and Engineering, Shanghai Jiao Tong University, Shanghai 200240, P. R. China

Dr. M. J. Shi

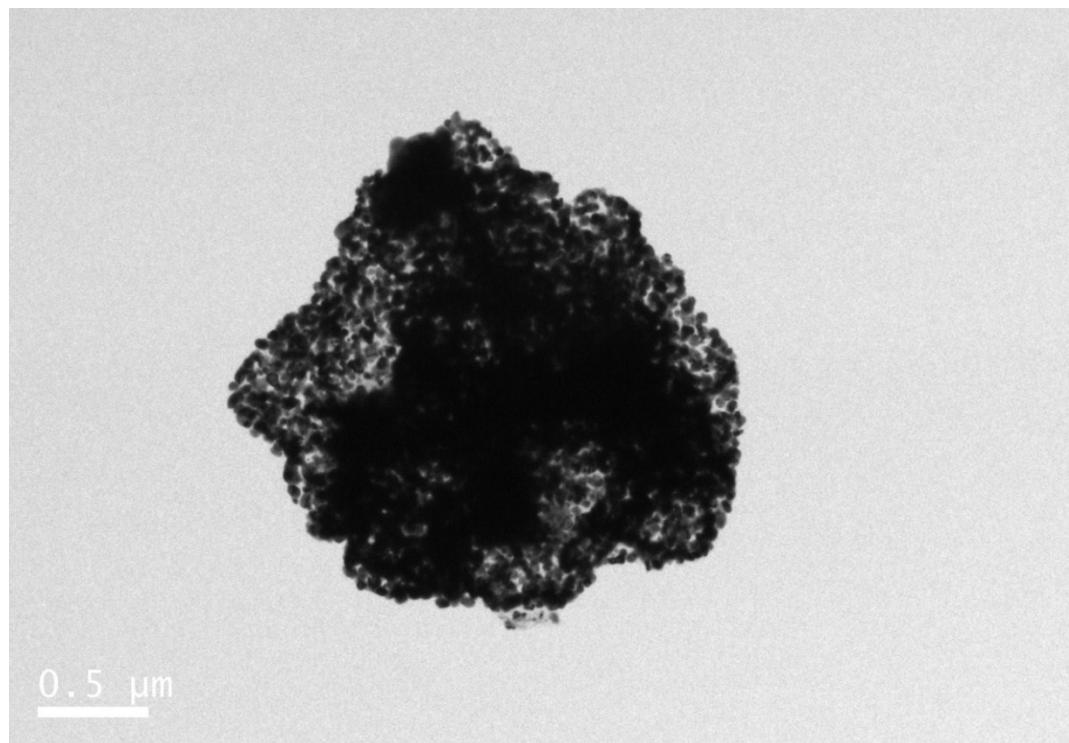
School of Materials Science and Engineering, Jiangsu University of Science and Technology, Zhenjiang, 212003, P. R. China.

Prof. J. Yang

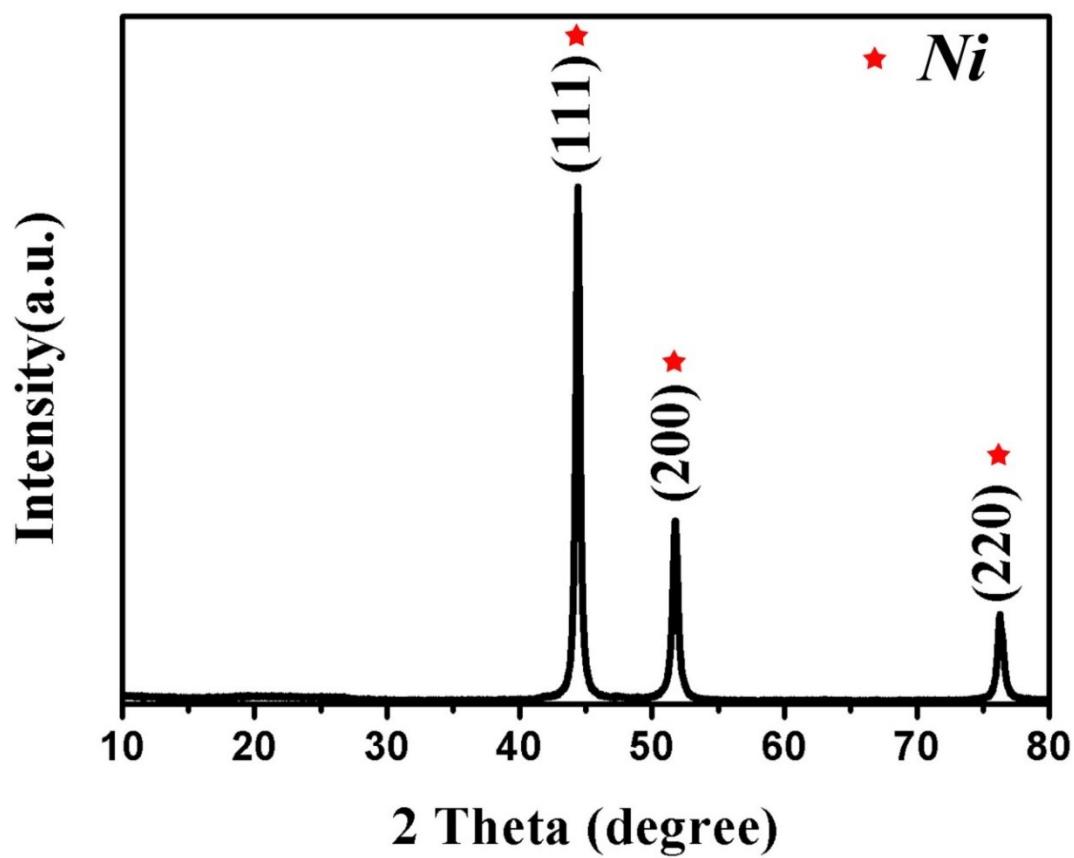
Shanghai Electrochemical Energy Devices Research Center, School of Chemistry and Chemical Engineering, Shanghai Jiao Tong University, Shanghai 200240, P. R. China



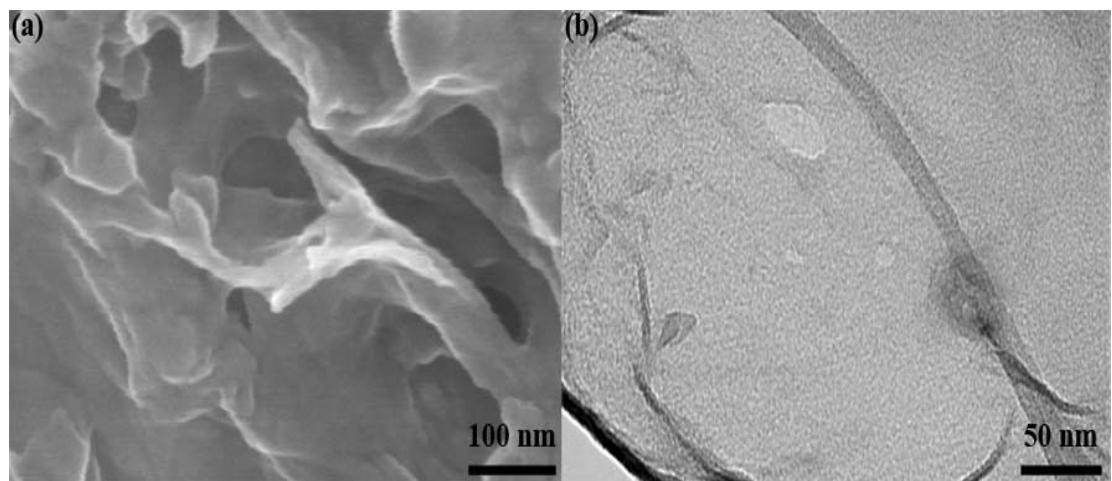
**Fig. S1.** The atomic force microscope (AFM) image of graphene oxide sheets. The thickness of graphene nanosheets is about 0.48 nm.



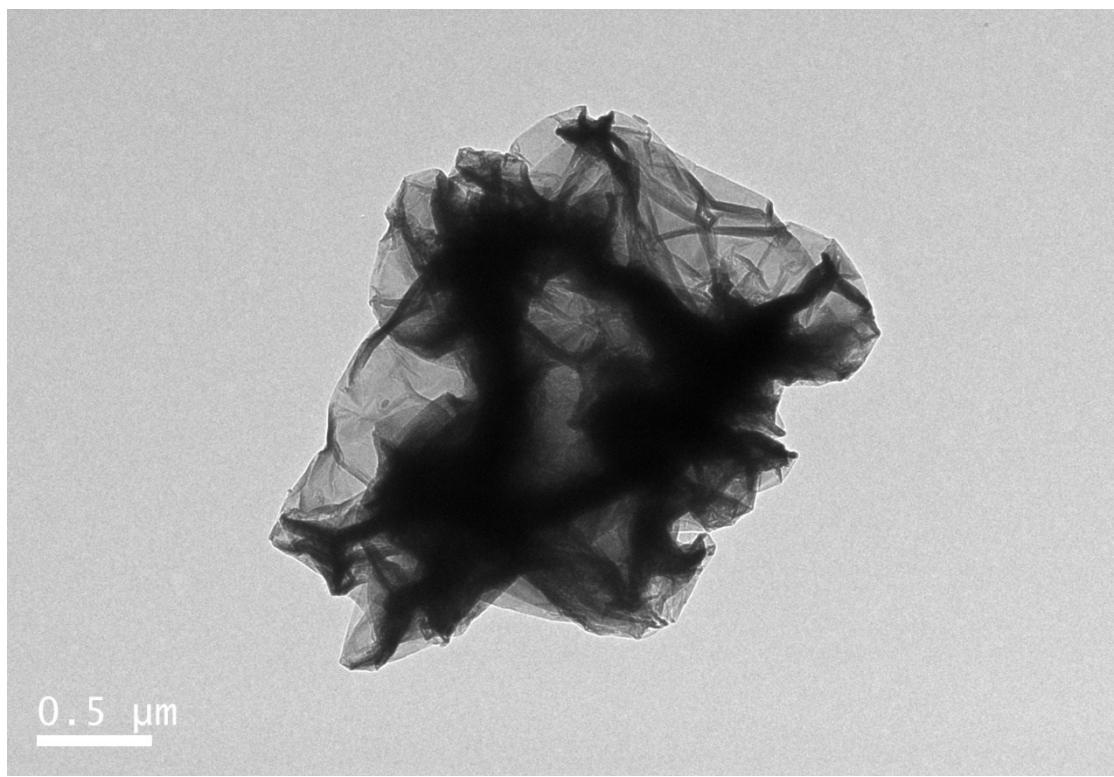
**Fig. S2.** TEM image of Ni@G.



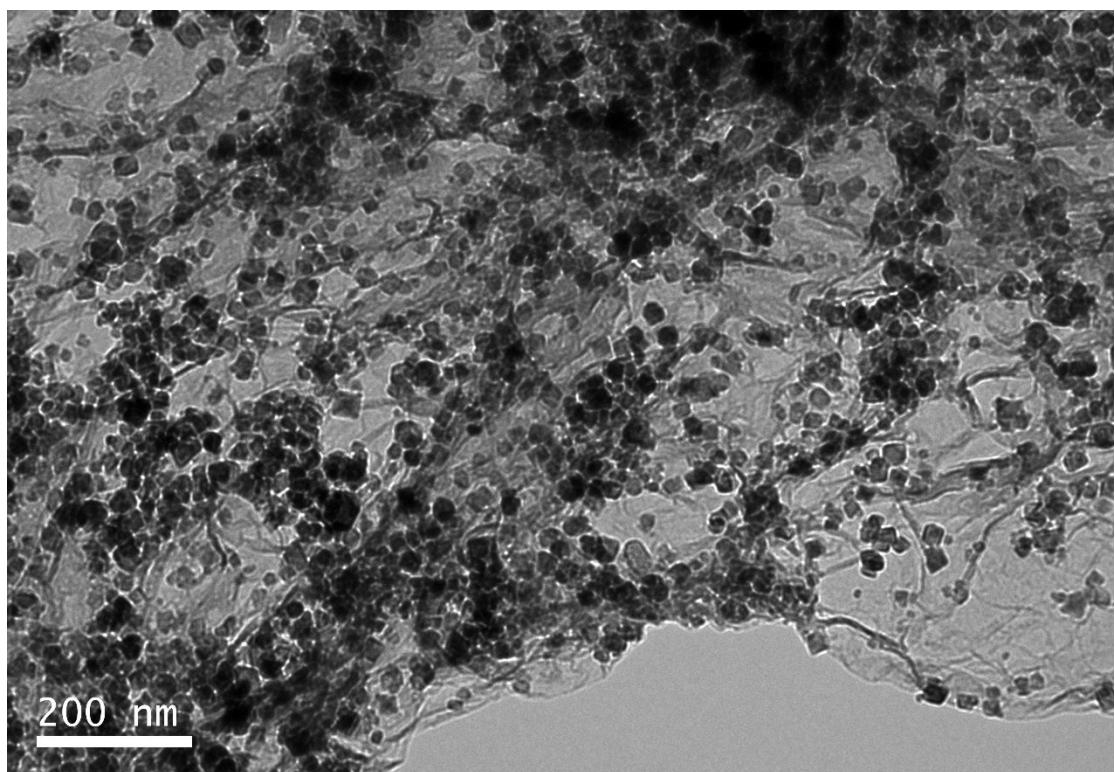
**Fig. S3.** XRD pattern of Ni@G.



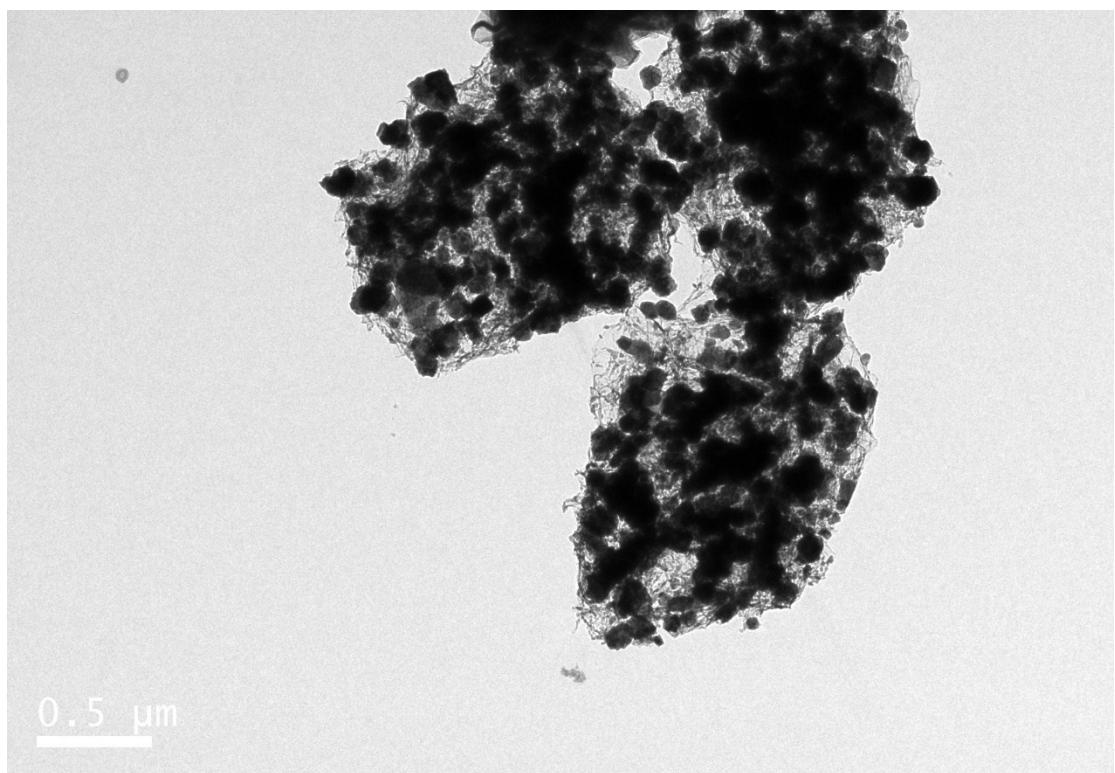
**Fig. S4.** (a) FESEM and (b) TEM images of porous graphene microspheres.



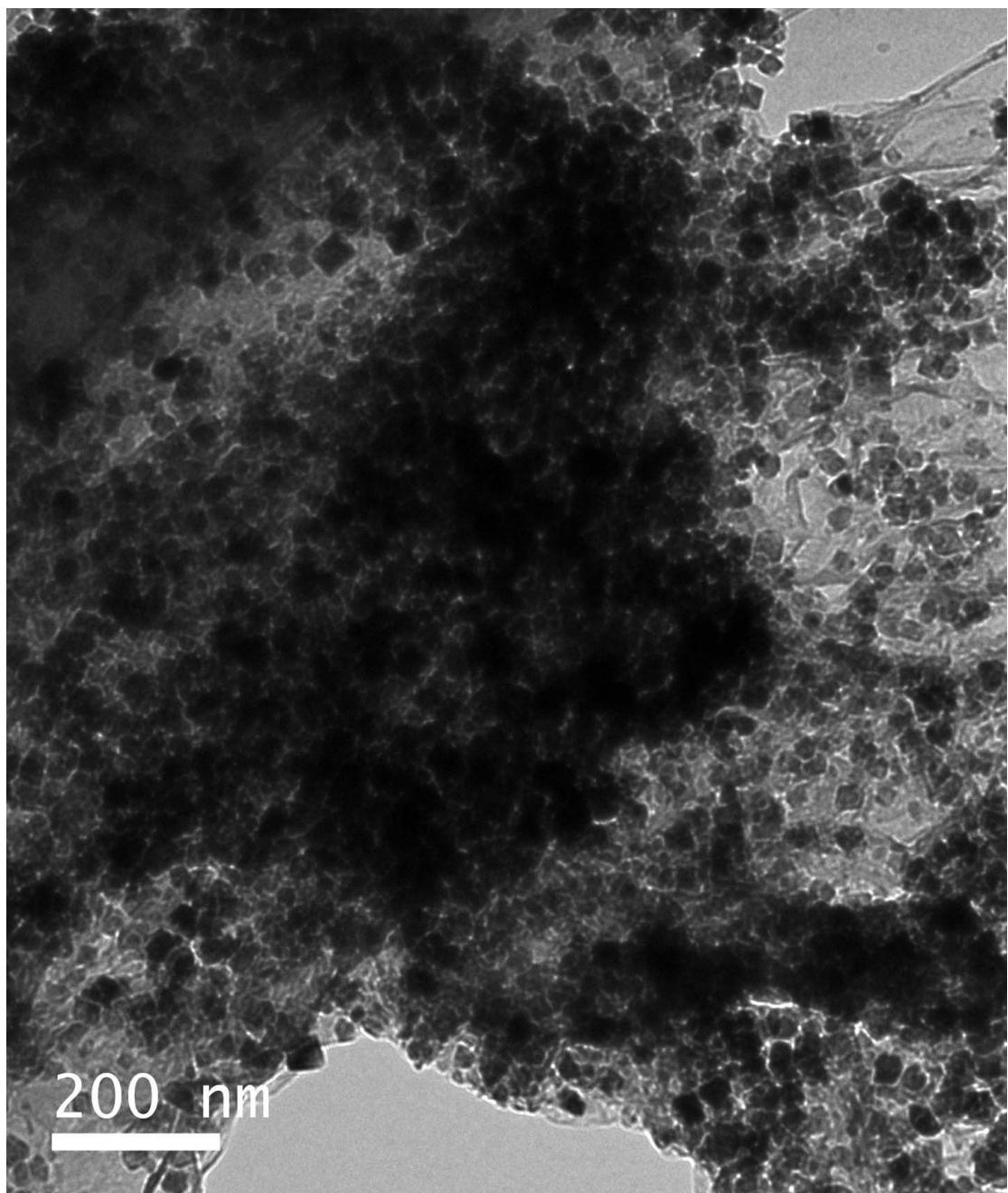
**Fig. S5.** TEM image of pristine graphene microspheres.



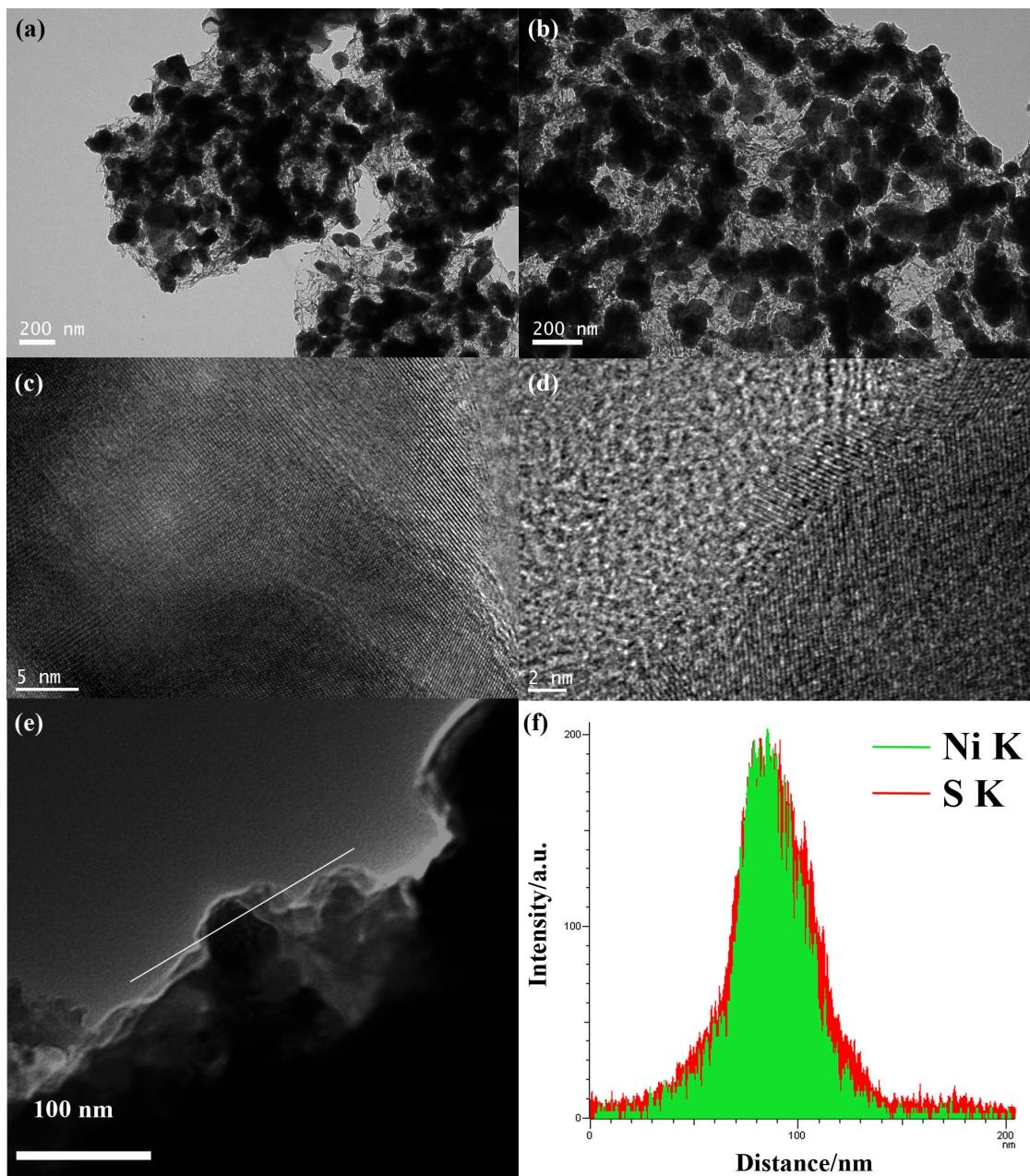
**Fig. S6.** TEM image of DM- $\text{Ni}_x\text{S}_y@\text{G}$ .



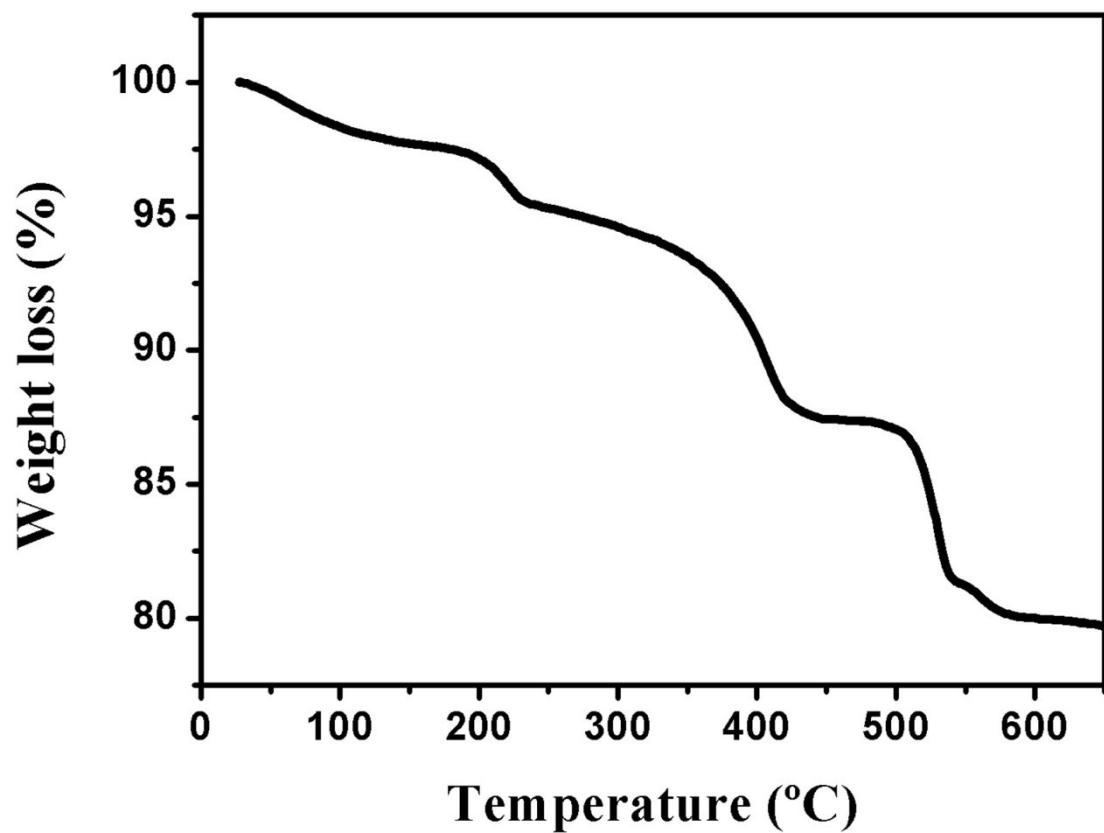
**Fig. S7.** TEM image of  $\text{NiS}@\text{G}$ .



**Fig. S8.** TEM image of DM-Ni<sub>x</sub>S<sub>y</sub>@G electrodes after 100000 charge-discharge cycles at 5 A/g.



**Fig. S9.** (a, b) TEM images, (c, d) HRTEM images of PM- $\text{Ni}_x\text{S}_y$  microflowers. (e) scanning transmission electron microscopy image for STEM/EDS line scan, (f) STEM/EDS line scan profile of binary  $\text{Ni}_x\text{S}_y$  nanobeads in PM- $\text{Ni}_x\text{S}_y$  microflowers.



**Fig. S10.** TGA curve of PM- $\text{Ni}_x\text{S}_y$  microflowers ranging from 25 to 650 °C at a heating rate of 10 °C /min under air flow.

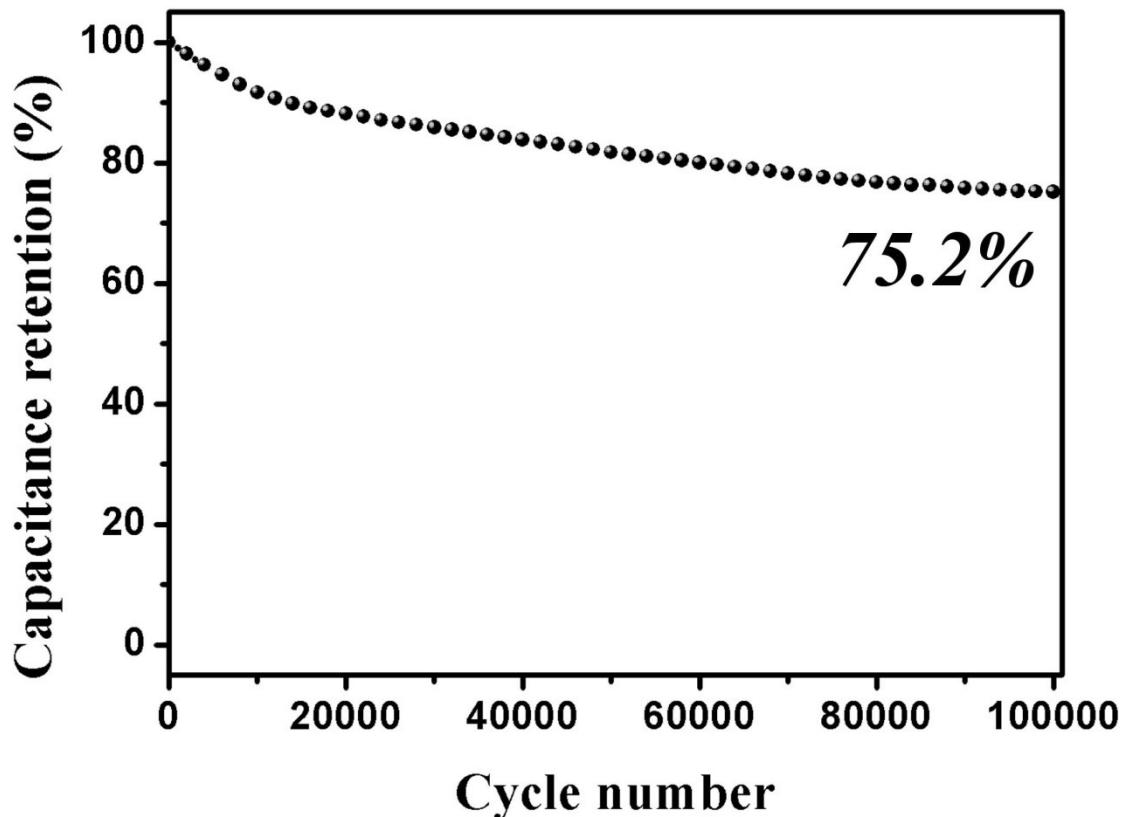


Fig. S11. Cycle performance of the DM- $\text{Ni}_x\text{S}_y@\text{G}$  electrodes.

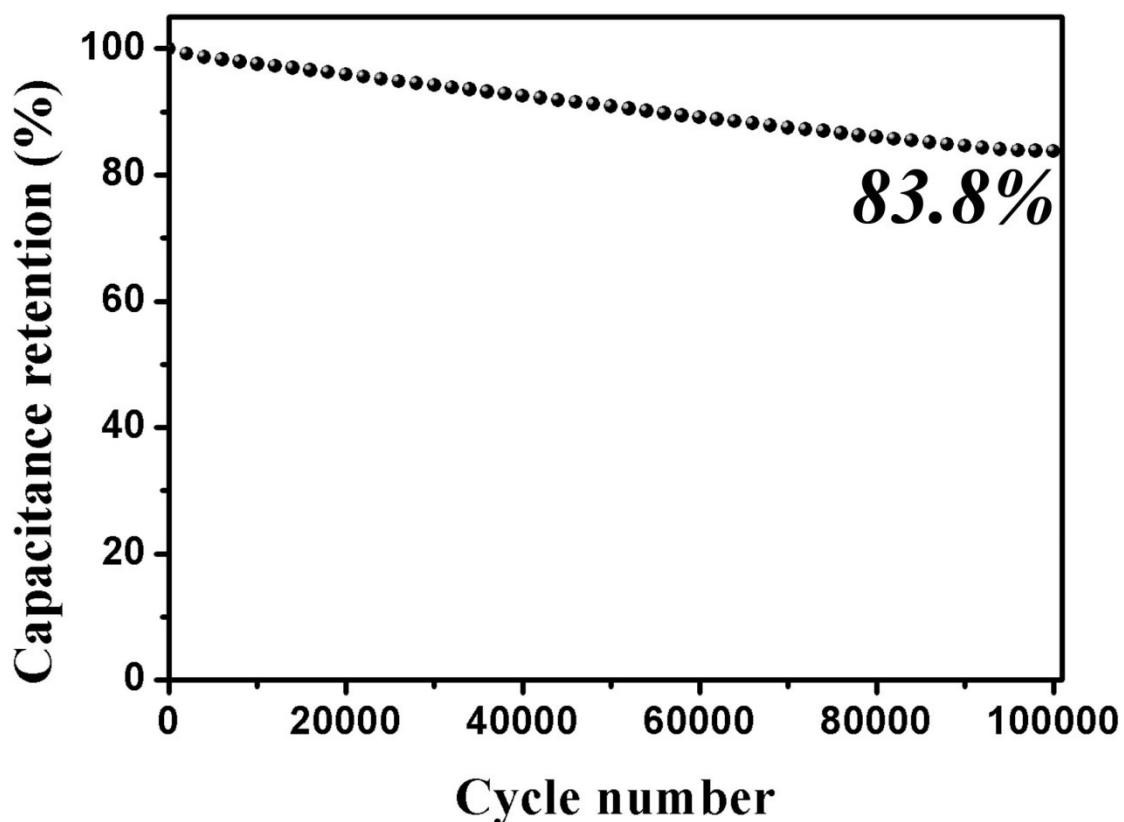
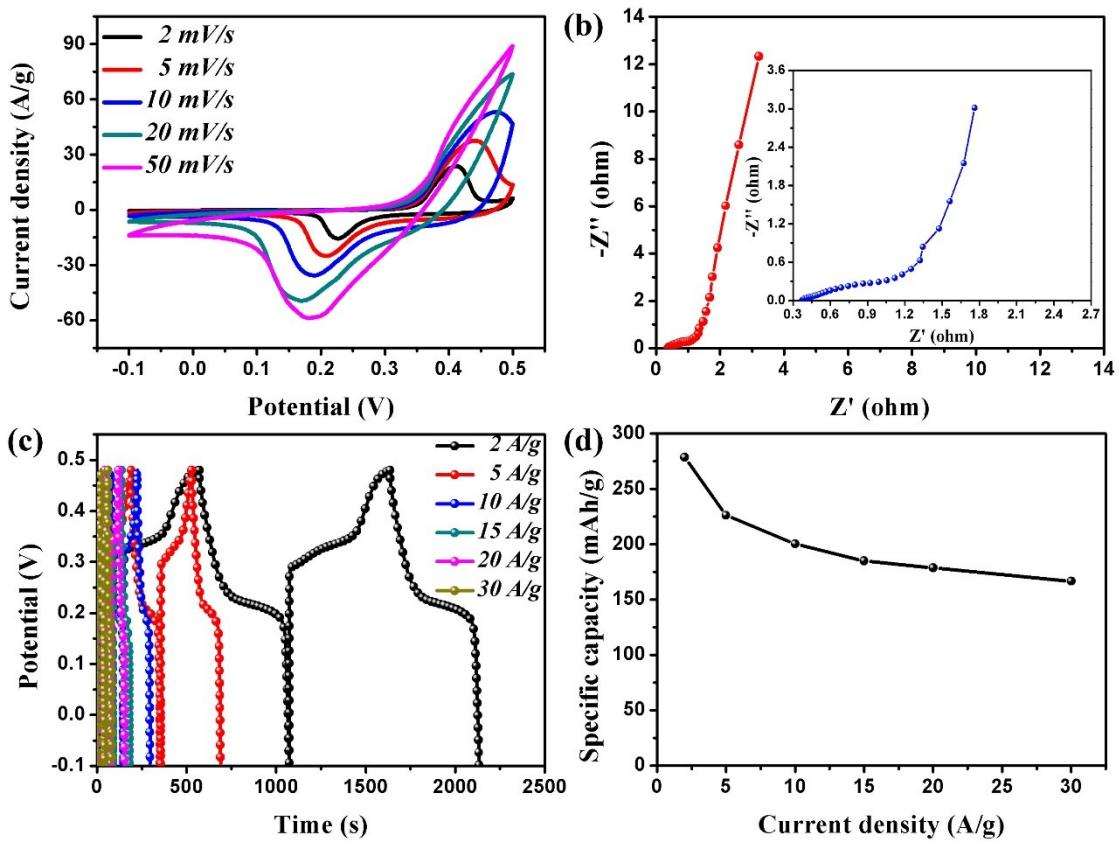
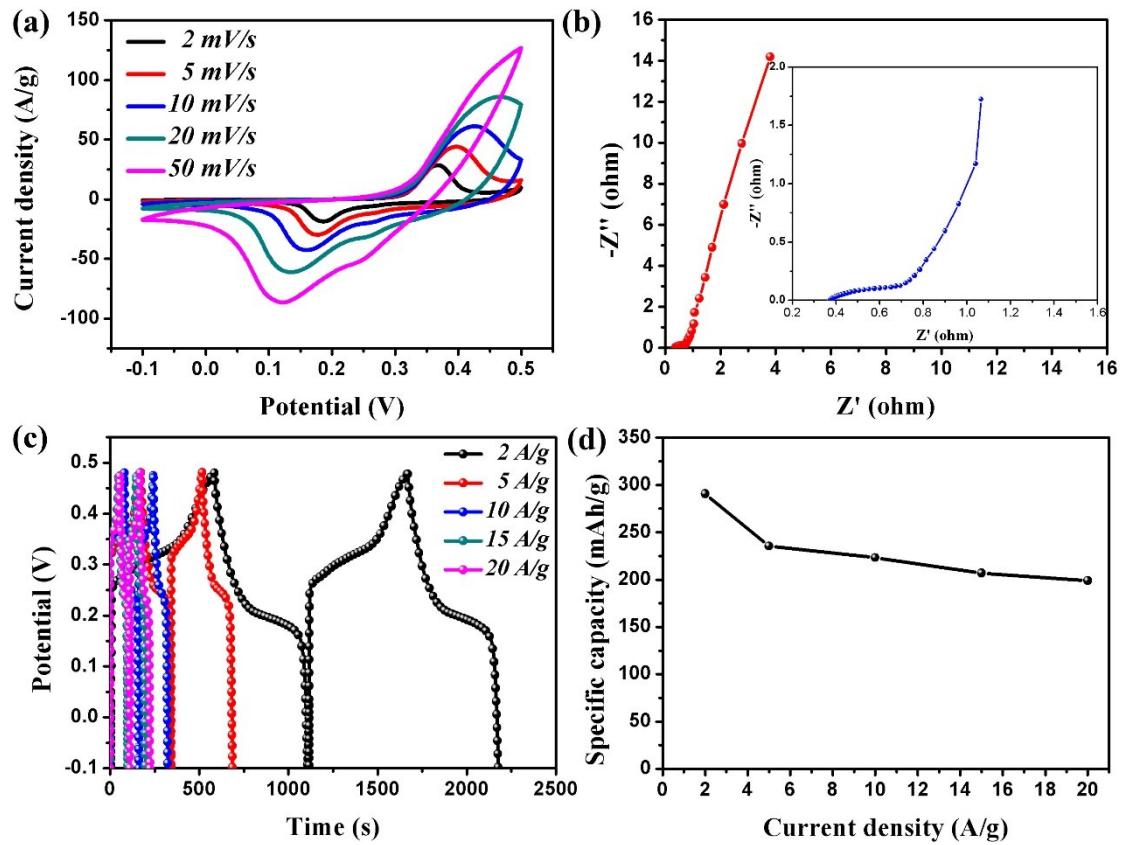


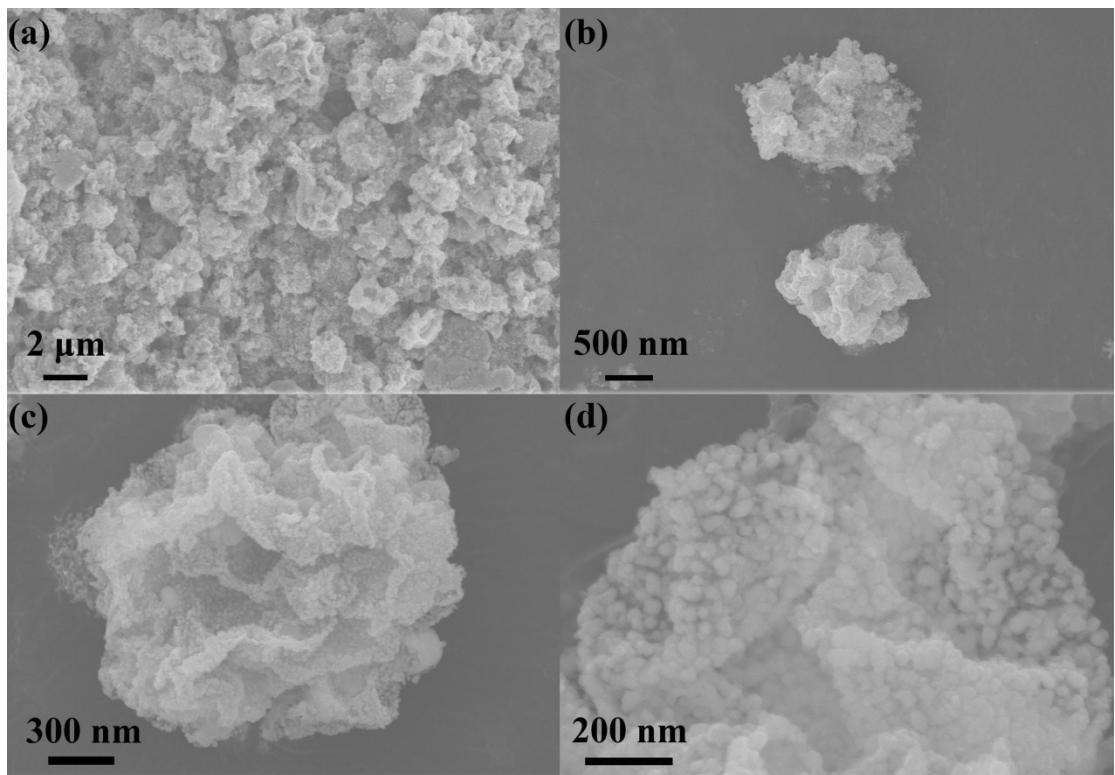
Fig. S12. Cycle performance of the  $\text{NiS}@\text{G}$  electrodes.



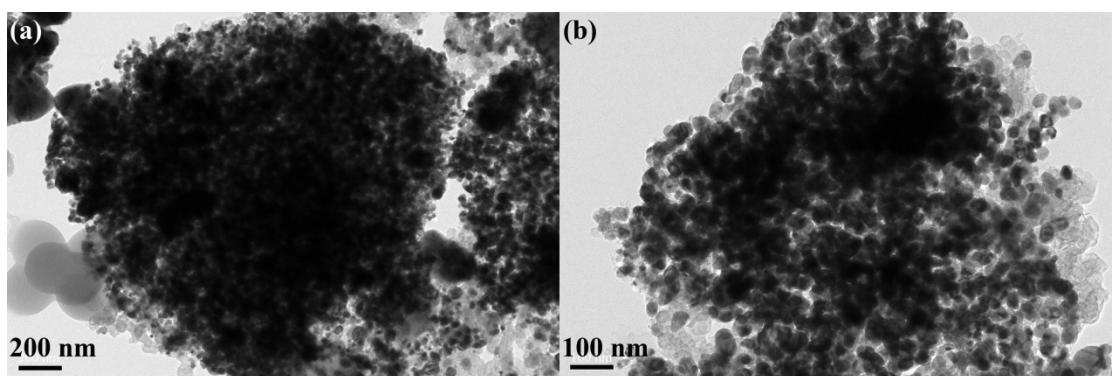
**Fig. S13.** (a) CV curves at scan rates of 2-50 mV/s; (b) EIS plot; (c) GCD curves; (d) specific capacity at different current densities of 2-30 A/g of DM-Ni<sub>x</sub>S<sub>y</sub>@G electrodes.



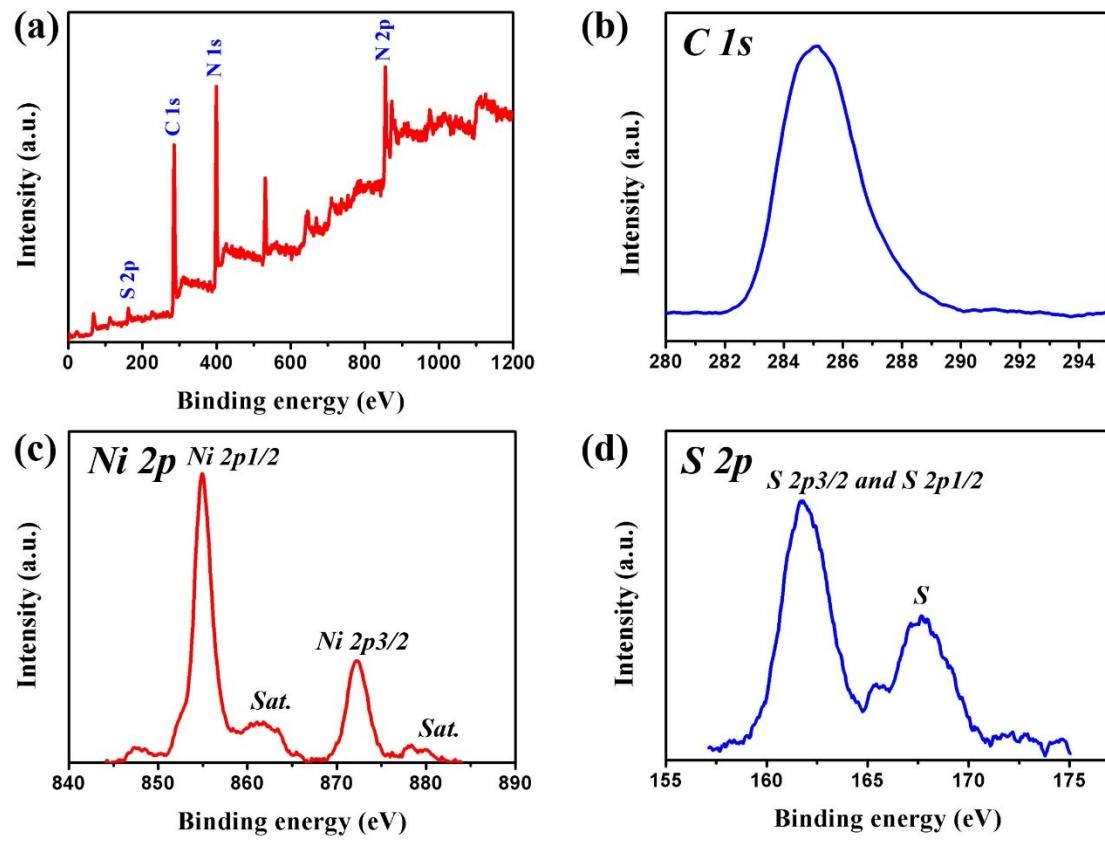
**Fig. S14.** (a) CV curves at scan rates of 2-50 mV/s; (b) EIS plot; (c) GCD curves; (d) specific capacity at different current densities of 2-20 A/g of NiS@G electrodes.



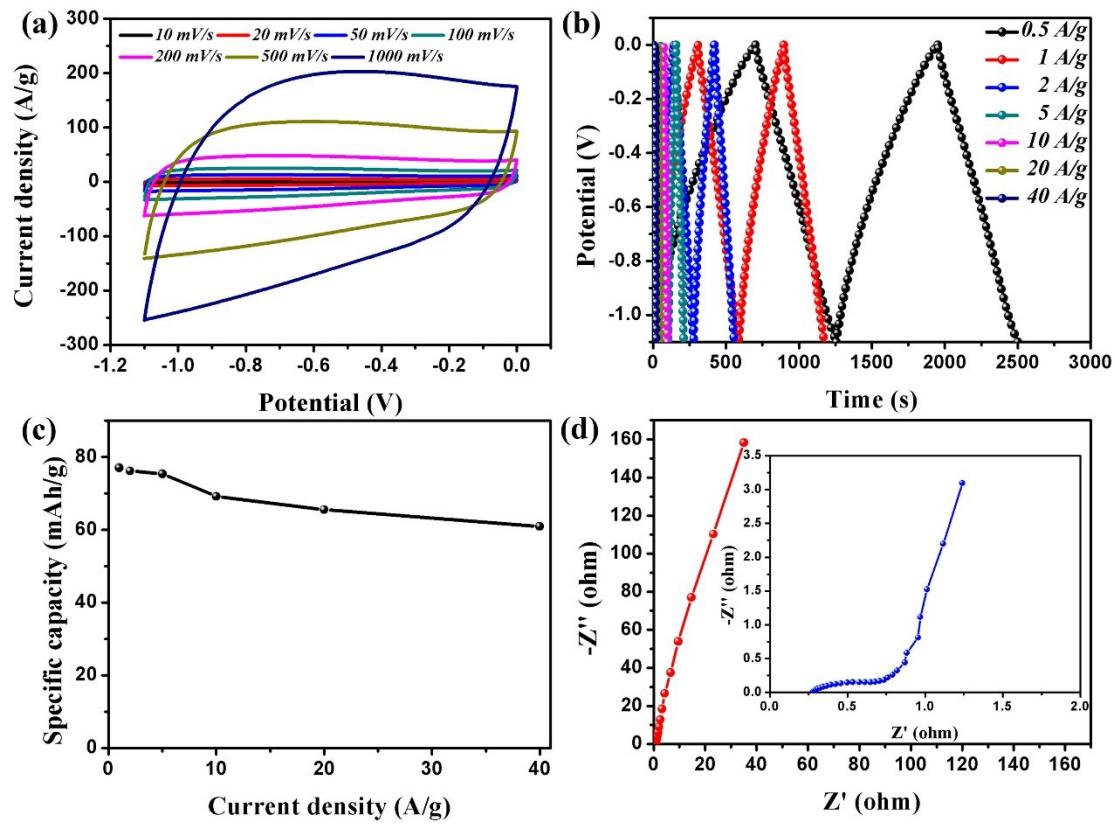
**Fig. S15.** SEM image of PM- $\text{Ni}_x\text{S}_y$  electrodes after 100 000 charge-discharge cycles at 5 A/g.



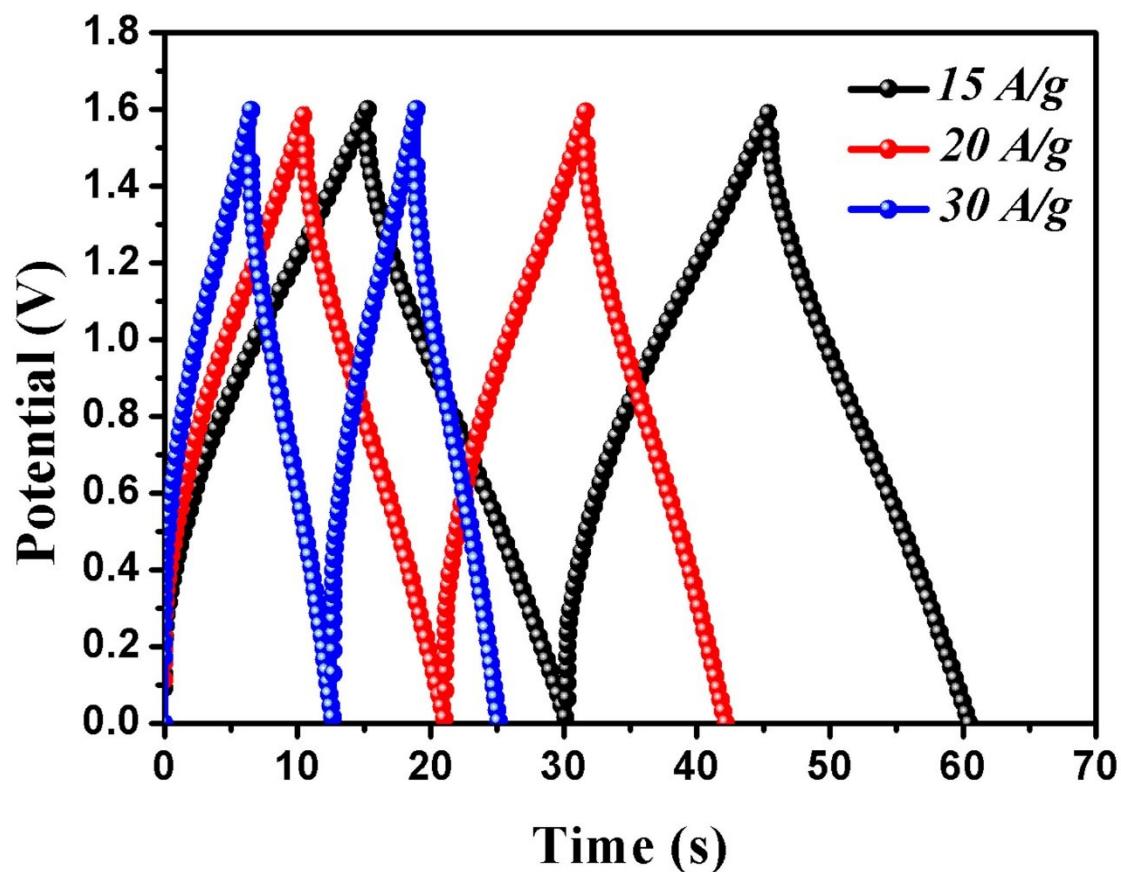
**Fig. S16.** TEM image of PM- $\text{Ni}_x\text{S}_y$  electrodes after 100 000 charge-discharge cycles at 5 A/g.



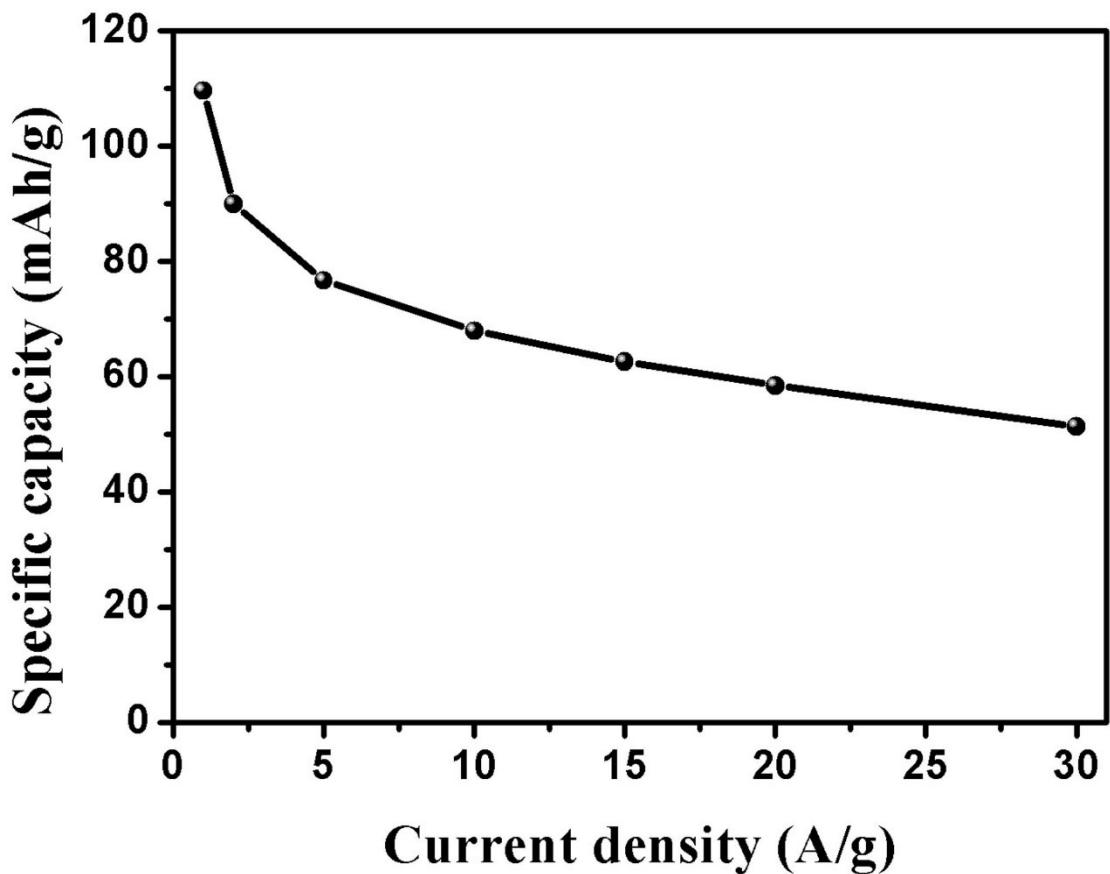
**Fig. S17.** XPS patterns of PM- $\text{Ni}_x\text{S}_y$  electrodes after 100 000 charge-discharge cycles at 5 A/g.



**Fig. S18.** (a) CV curves at scan rates of 10- 1000 mV/s; (b) GCD curves; (c) specific capacity at different current densities; (d) EIS plot of porous graphene microspheres.



**Fig. S19.** GCD curves at different current densities of 15-30 A/g of PM-Ni<sub>x</sub>S<sub>y</sub>//PG ASCs.



**Fig. S20.** Specific capacity at different current densities of 1-30 A/g of PM- $\text{Ni}_x\text{S}_y/\text//\text{PG}$  ASCs.

**Table S1.** Comparision study of the cyclic stability between the PM-Ni<sub>x</sub>S<sub>y</sub> microflowers electrode in this work and previously reported nickel sulfides based materials.

Materials	Capacitance (F/g)	Cyclic stability	Ref
Ni@rGO-Ni <sub>3</sub> S <sub>2</sub>	987.8 (1.5 A/g)	97.9% (12 A/g, 3000 cycles)	[1]
Rod-like NiS <sub>2</sub>	1020.2 (1 A/g )	93.4% (2 A/g, 2000 cycles)	[2]
NiS microflowers	1122.7 (1 A/g )	97.8% (10 A/g, 1000 cycles)	[3]
Ni <sub>3</sub> S <sub>4</sub> -MoS <sub>2</sub> heterojunction	985.21 (1 A/g )	85% (10 A/g, 20000 cycles)	[4]
NiS <sub>2</sub> /ZnS	1198 (1 A/g )	87% (5 A/g, 1000 cycles)	[5]
NiS/Ni <sub>3</sub> S <sub>4</sub> nanosheets	2070.0 (2.5 A/g )	86% (3 A/g, 10000 cycles)	[6]
Ni <sub>3</sub> S <sub>2</sub> /MWCNT	1024 (0.8 A/g )	80% (3.2 A/g, 1000 cycles)	[7]
Ni <sub>3</sub> S <sub>4</sub> @MoS <sub>2</sub>	1440.9 (2 A/g )	90.7% (10 A/g, 3000 cycles)	[8]
Honeycomb-like NiS <sub>2</sub> /NiO	2251 (1 A/g )	78% (5 A/g, 2000 cycles)	[9]
Co-Mo-O/Ni <sub>3</sub> S <sub>2</sub> /NF	340 (2 A/g )	95.7% (2 A/g, 2000 cycles)	[10]
Co-doped NiS	2141.9 (2 A/g )	75.3% (10 A/g, 3000 cycles)	[11]
NiCo <sub>2</sub> S <sub>4</sub> /carbon sponge	1093 (0.5 A/g )	83% (8A/g, 8000 cycles)	[12]
Ni <sub>x</sub> S <sub>y</sub> @CoS polyhedrons	2091.4 (2 A/g )	85.2% (5 A/g, 2000 cycles)	[13]
NiS <sub>2</sub> /CoS <sub>2</sub>	954.3 (1 A/g )	99.9% (5 A/g, 1000 cycles)	[14]
NiS <sub>x</sub> @NCV microcapsules	1600 (1 A/g )	94.36% (1 A/g, 5000 cycles)	[15]
PM-Ni <sub>x</sub> S <sub>y</sub> microflowers	2331.3 (323.8 mAh/g, 2 A/g)	98.9% (5 A/g, 100000 cycles)	This work

**Table S2.** Comparision study of the cyclic stability, energy density and power density

between the PM-Ni<sub>x</sub>S<sub>y</sub>//PG ASCs in this work and previously reported nickel sulfides based SCs.

Device	Cyclic stability	Energy density (W h/kg)	Power density (kW/kg)	Ref
Ni@rGO-Co <sub>3</sub> S <sub>4</sub> //Ni@rGO-Ni <sub>3</sub> S <sub>2</sub>	96.2% after 3000 cycles (12 A/g)	55.16	0.975	[1]
Rod-like NiS <sub>2</sub> //rGO	84.1% after 4000 cycles (2 A/g)	32.76	0.954	[2]
NiS-NF//AC	90.6% after 1000 cycles (25 mA/cm <sup>2</sup> )	38.4	0.1666	[16]
Ni <sub>3</sub> S <sub>4</sub> -MoS <sub>2</sub> heterojunction//AC	86.2% after 10000 cycles (10 A/g)	58.43	0.38595	[3]
Co-Mo-O/Ni <sub>3</sub> S <sub>2</sub> /NF	90.2% after 3000 cycles (12 A/g)	57.9	1.1	[10]
Co-doped NiS//AC	85.1% after 4000 cycles (10 A/g)	54.9	1.5154	[11]
NiS <sub>2</sub> -CoS <sub>2</sub> //AC	99.1% after 4000 cycles (2 A/g)	29.3	0.7	[14]
Ni <sub>3</sub> S <sub>2</sub> /MWCNT-NC//AC	90% after 5000 cycles (4 A/g)	19.8	0.798	[7]
Ni <sub>3</sub> S <sub>2</sub> @β-NiS//AC	97.8% after 10000 cycles (5 A/g)	55.1	0.9259	[17]
NiS <sub>2</sub> /ZnS//AC	88.7% after 1500 cycles (5 A/g)	28	0.7489	[5]
Ni <sub>3</sub> S <sub>2</sub> /CNF//CNF	97% after 2500 cycles (100 mV/s)	25.8	0.425	[18]
R-NiS/rGO//C/NG-A	93.2% after 10000 cycles (20 A/g)	93	0.962	[19]
NiS <sub>x</sub> @NCV//NPC	92.4% after 5000 cycles (1 A/g)	48.02	0.8	[15]
PM-Ni <sub>x</sub> S <sub>y</sub> //PG	97.6% after 100000 cycles (5 A/g)	88	0.81	This work

**Table S3.** The rated voltage and power of the used equipment in this work.

Equipment	Rated voltage/V	Rated power/W
LED lamp	1.5	3
Electronic motor	2	5
Miniature game machine	1.5	4
Colorful illuminations	3	5
Electronic game machine	3	7

## Notes and references

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