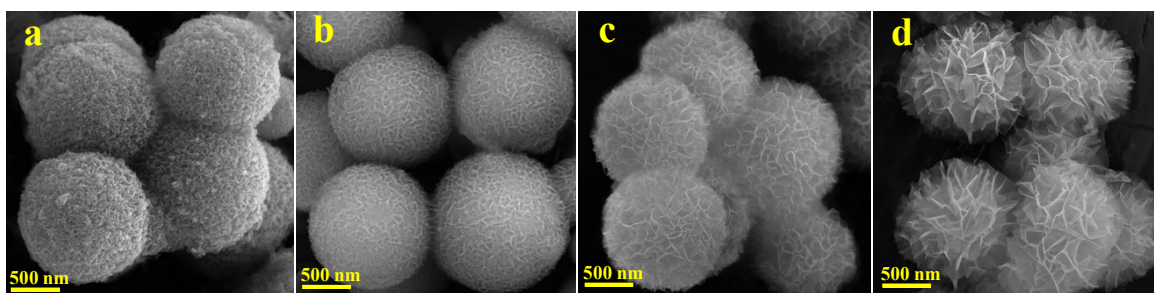


## Supporting Information

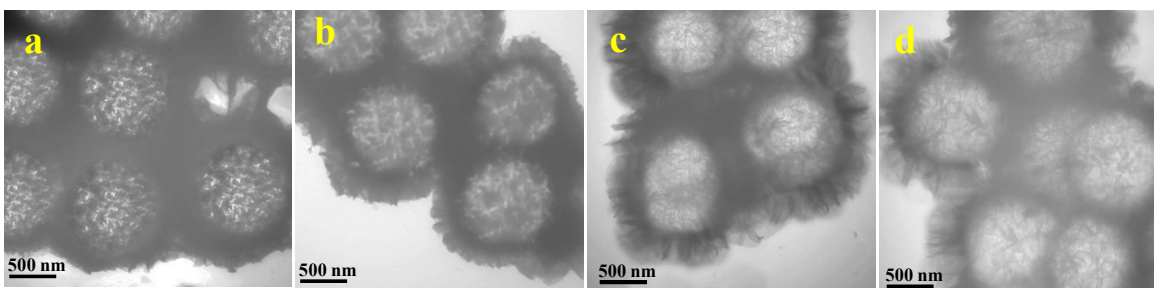
### Fabrication of nanosheet-assembled hollow copper-nickel phosphide spheres embedded in reduced graphene oxide texture for hybrid supercapacitors

Maryam Amiri, Akbar Mohammadi Zardkhoshoui\* and Saied Saeed Hosseiny Davarani\*

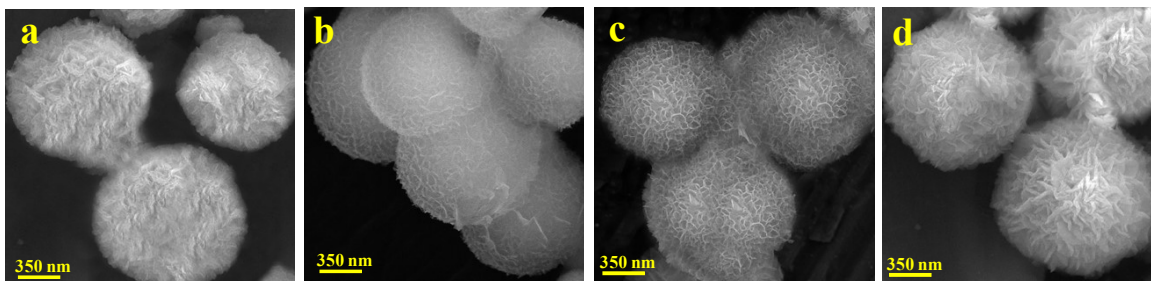
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Corresponding authors: \*Tel: +98 21 22431661; Fax: +98 21 22431661; E-mail: [ss-hosseiny@sbu.ac.ir](mailto:ss-hosseiny@sbu.ac.ir) (S.S.H. Davarani); and [mohammadi.bahadoran@gmail.com](mailto:mohammadi.bahadoran@gmail.com) (A. Mohammadi Zardkhoshoui)



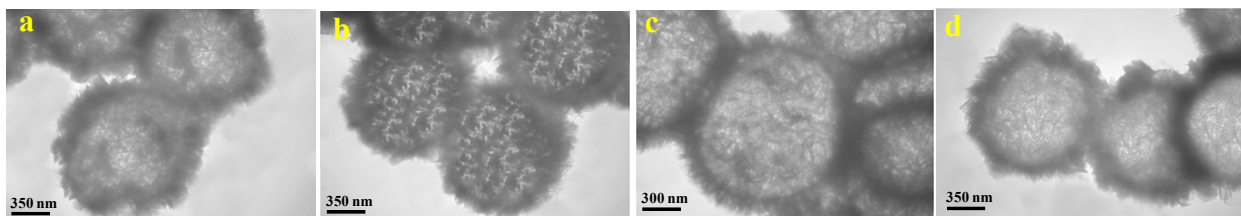
**Fig. S1** (a) FE-SEM image of the CN-EG32-100. (b) FE-SEM image of the CN-EG32-130. (c) FE-SEM image of the CN-EG32-160. (d) FE-SEM image of the CN-EG32-180.



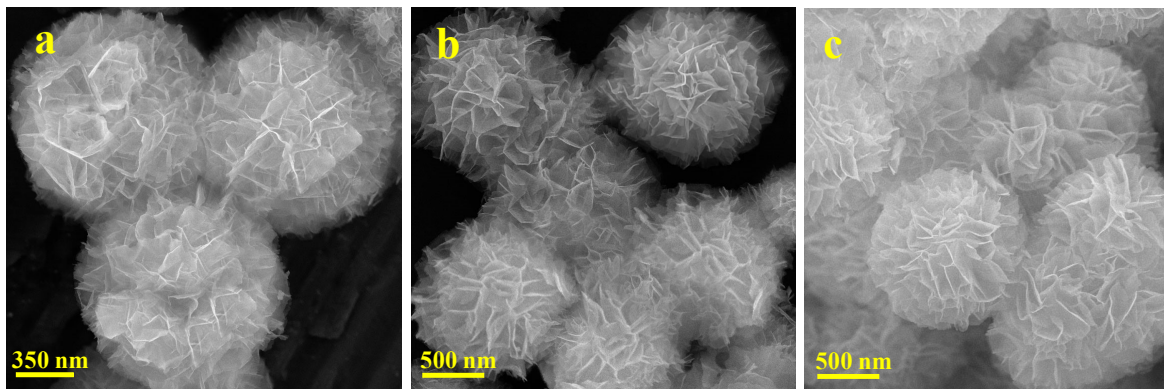
**Fig. S2** (a) TEM image of the CN-EG32-100. (b) TEM image of the CN-EG32-130. (c) TEM image of the CN-EG32-160. (d) TEM image of the CN-EG32-180.



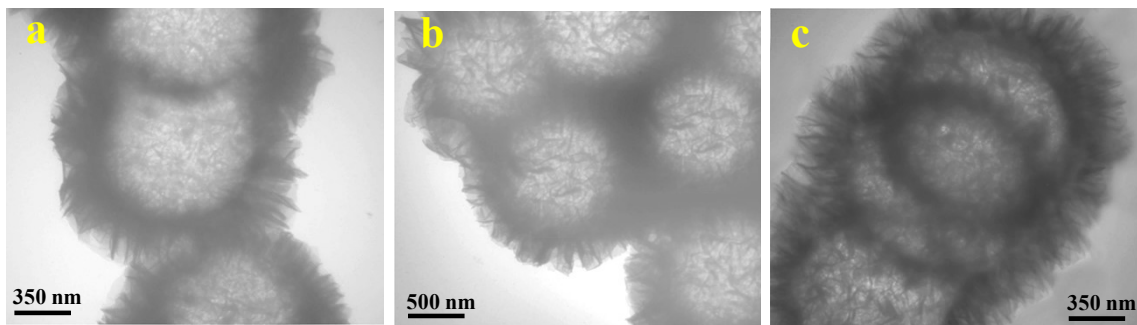
**Fig. S3** (a) FE-SEM image of the CN-EG11-180. (b) FE-SEM image of the CN-EG12-180. (c) FE-SEM image of the CN-EG21-180. (d) FE-SEM image of the CN-EG23-180.



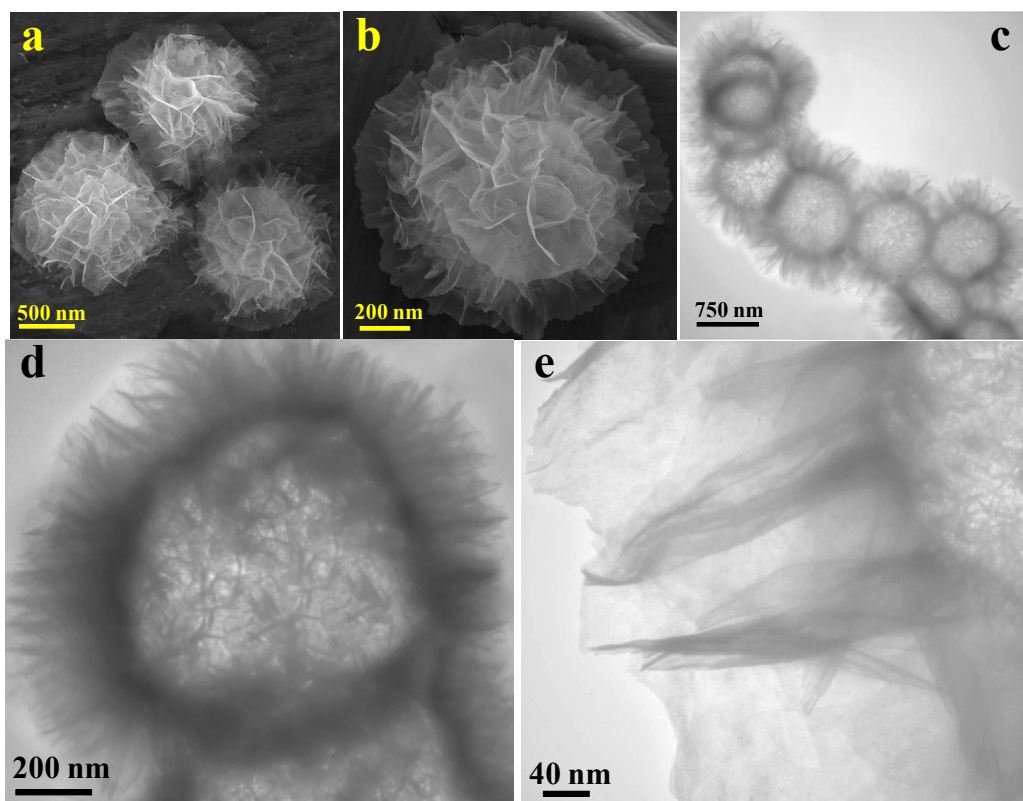
**Fig. S4** (a) TEM image of the CN-EG11-180. (b) TEM image of the CN-EG12-180. (c) TEM image of the CN-EG21-180. (d) TEM image of the CN-EG23-180.



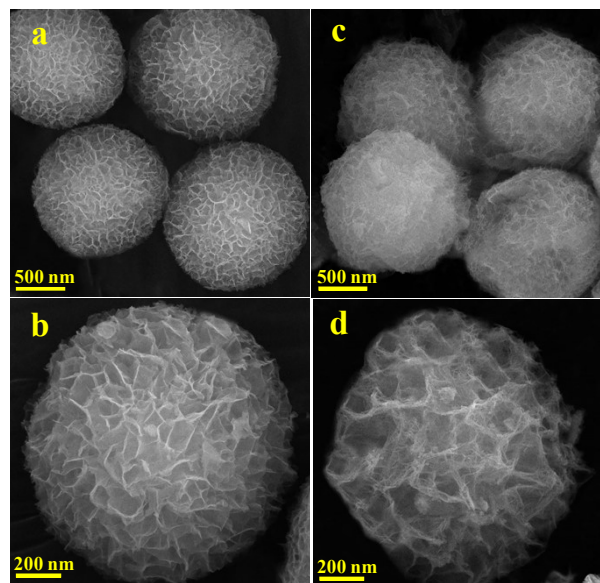
**Fig. S5** (a) FE-SEM image of the NH-CNPS-150. (b) FE-SEM image of the NH-CNPS-250. (c) FE-SEM image of the NH-CNPS-450.



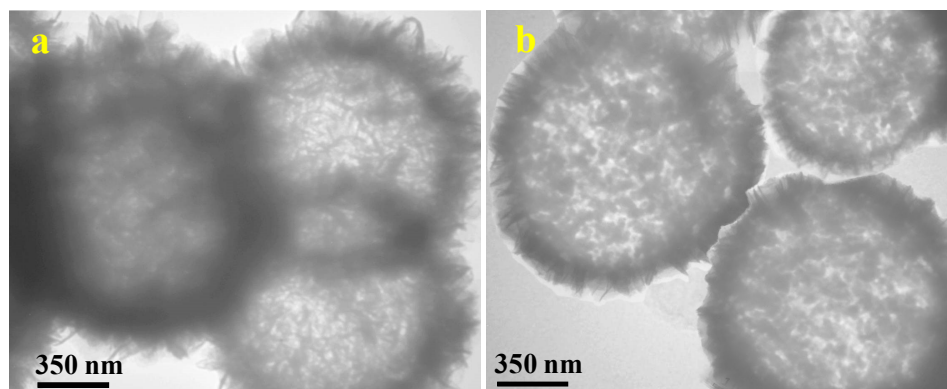
**Fig. S6** (a) TEM image of the NH-CNPS-150. (b) TEM image of the NH-CNPS-250. (c) TEM image of the NH-CNPS-450.



**Fig. S7** (a, b) FE-SEM images of the NH-CNPS-350. (c-e) TEM images of the NH-CNPS-350.



**Fig. S8** (a, b) FE-SEM images of the single Cu<sub>3</sub>P. (c, d) FE-SEM images of the single Ni<sub>2</sub>P.



**Fig. S9** (a) TEM image of the single Cu<sub>3</sub>P. (b) FE-SEM image of the single Ni<sub>2</sub>P.

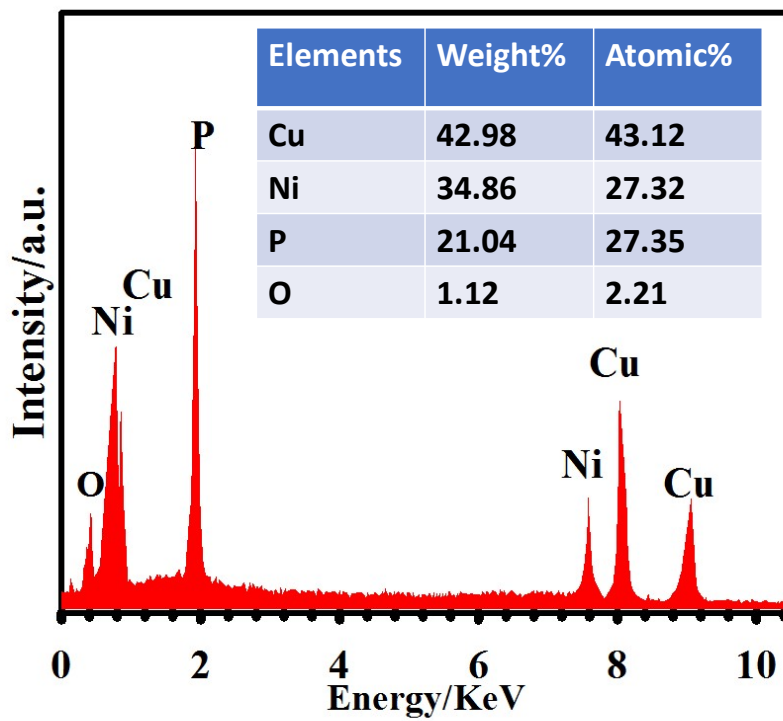


Fig. S10 EDX pattern of the NH-CNPS-350.

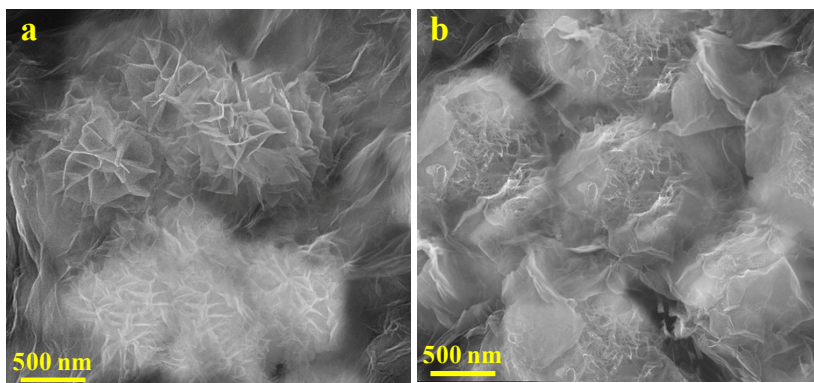
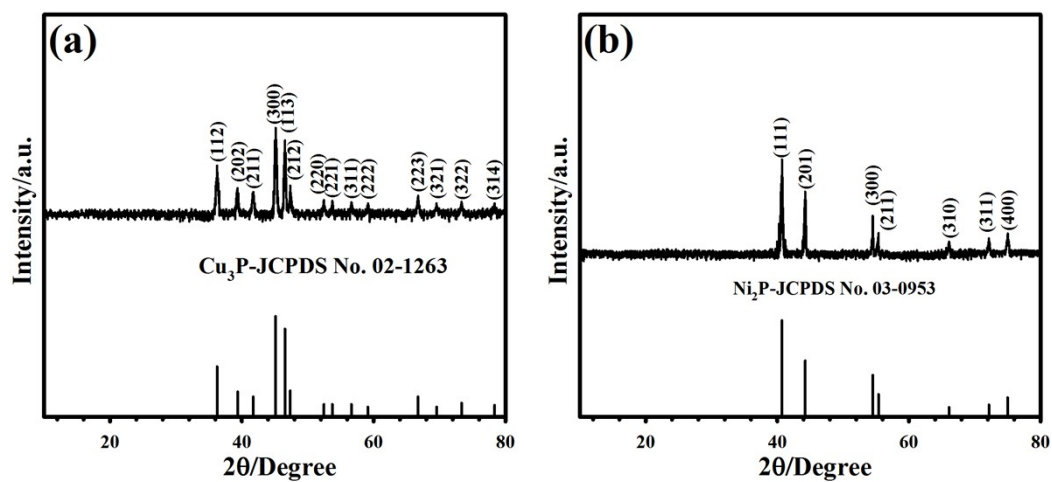
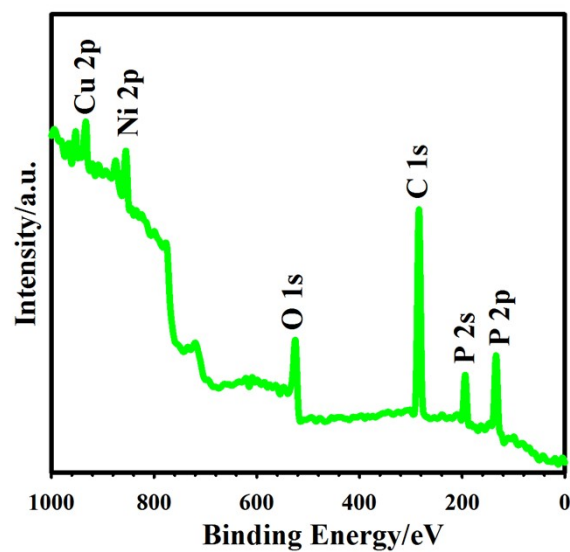


Fig. S11 (a) FE-SEM image of the NH-CNPS-rGO-1. (b) FE-SEM image of the NH-CNPS-rGO-3.

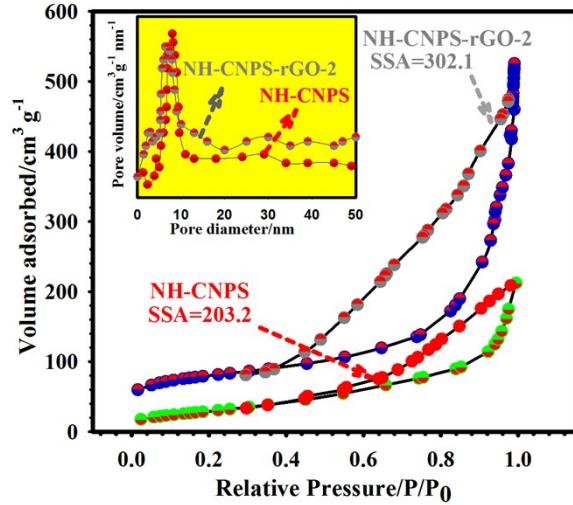




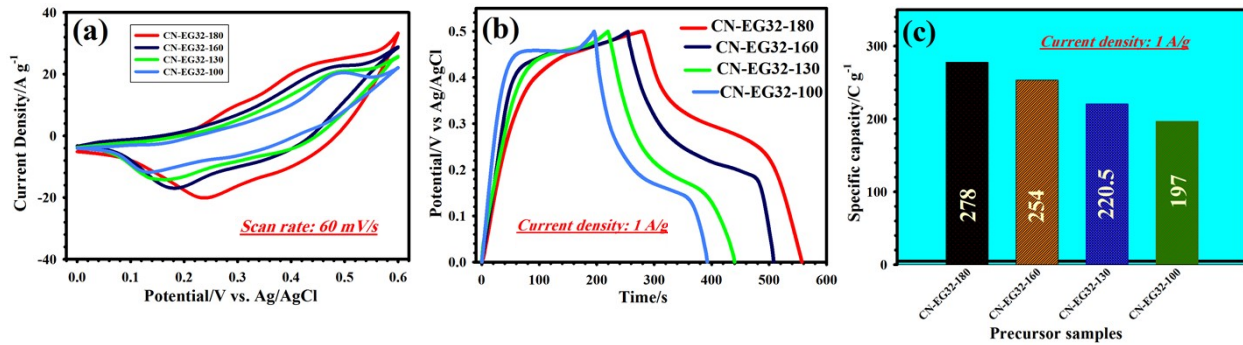
**Fig. S12** (a) XRD pattern of the  $\text{Cu}_3\text{P}$ . (b) XRD pattern of the  $\text{Ni}_2\text{P}$ .



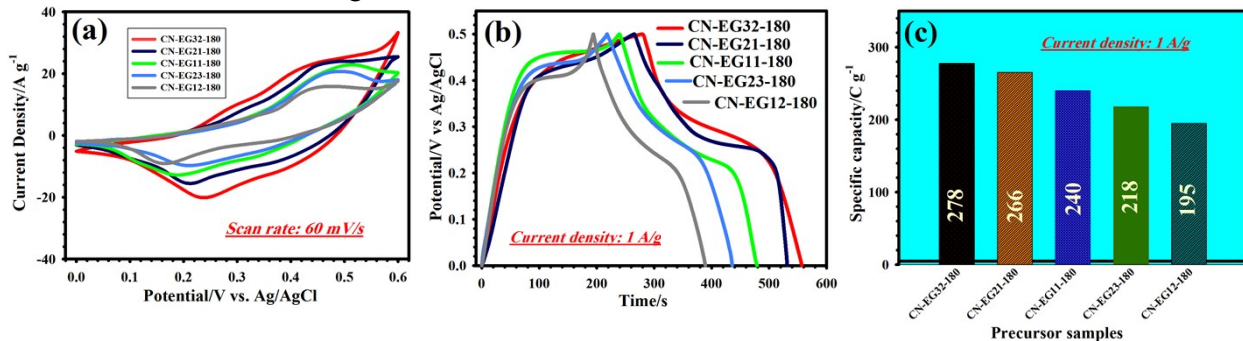
**Fig. S13** Survey profile of NH-CNPS-rGO-2.



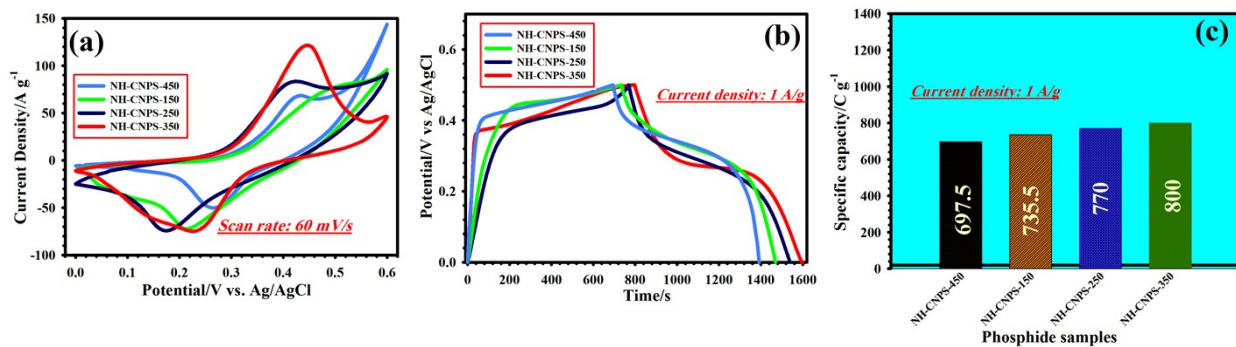
**Fig. S14** (h) BET plots of the NH-CNPS-350 and NH-CNPS-rGO-2 samples and their corresponding BJH curves (inset).



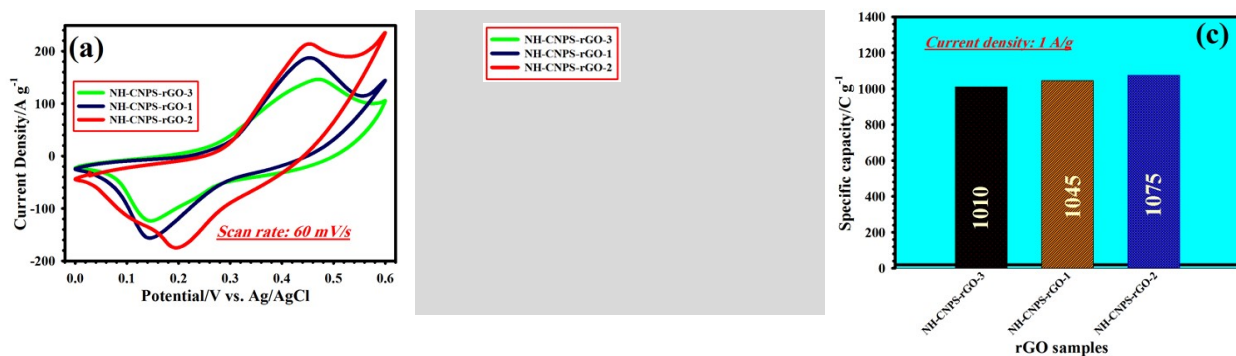
**Fig. 15** (a) CV curves of the CN-EG32-180, CN-EG32-160, CN-EG32-130, and CN-EG32-100 electrodes at 60 mV/s. (b) GCD curves of the CN-EG32-180, CN-EG32-160, CN-EG32-130, and CN-EG32-100 electrodes at 1 A/g. (c) Specific capacities of CN-EG32-180, CN-EG32-160, CN-EG32-130, and CN-EG32-100 electrodes at 1 A/g.



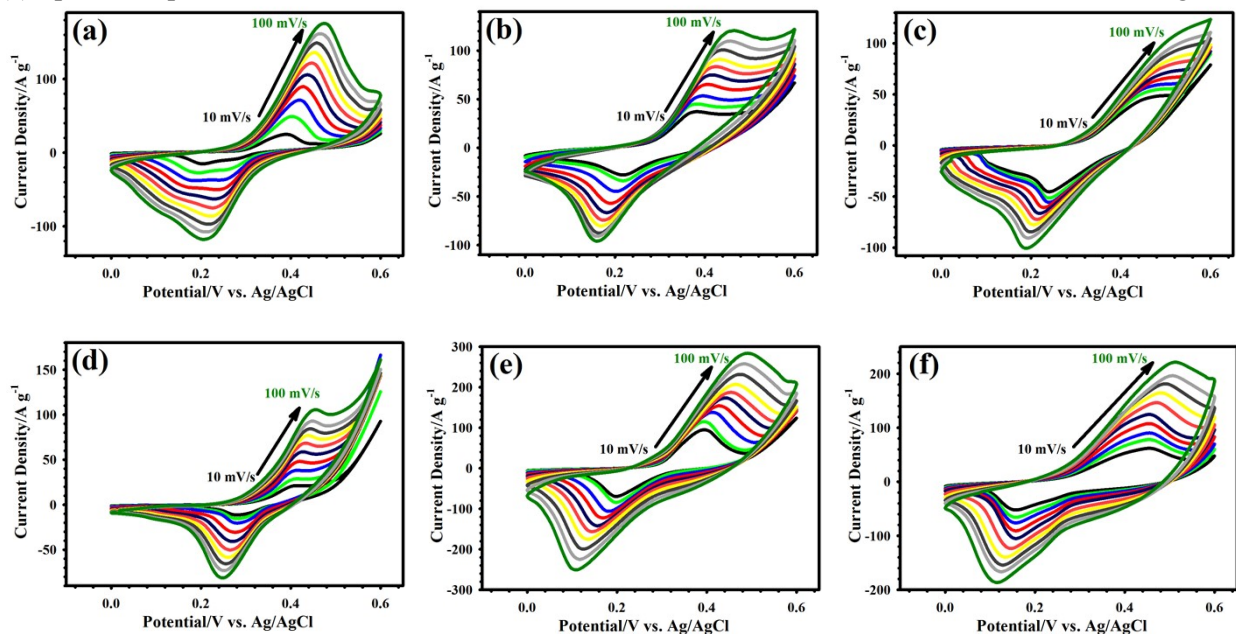
**Fig. 16** (a) CV curves of the CN-EG32-180, CN-EG21-180, CN-EG11-180, CN-EG23-180, and CN-EG12-180 electrodes at 60 mV/s. (b) GCD curves of the CN-EG32-180, CN-EG21-180, CN-EG11-180, CN-EG23-180, and CN-EG12-180 electrodes at 1 A/g. (c) Specific capacities of CN-EG32-180, CN-EG21-180, CN-EG11-180, CN-EG23-180, and CN-EG12-180 electrodes at 1 A/g.



**Fig. 17** (a) CV curves of the NH-CNS-150, NH-CNS-250, NH-CNS-350, and NH-CNS-450 electrodes at 60 mV/s. (b) GCD curves of the NH-CNS-150, NH-CNS-250, NH-CNS-350, and NH-CNS-450 electrodes at 1 A/g. (c) Specific capacities of NH-CNS-150, NH-CNS-250, NH-CNS-350, and NH-CNS-450 electrodes at 1 A/g.

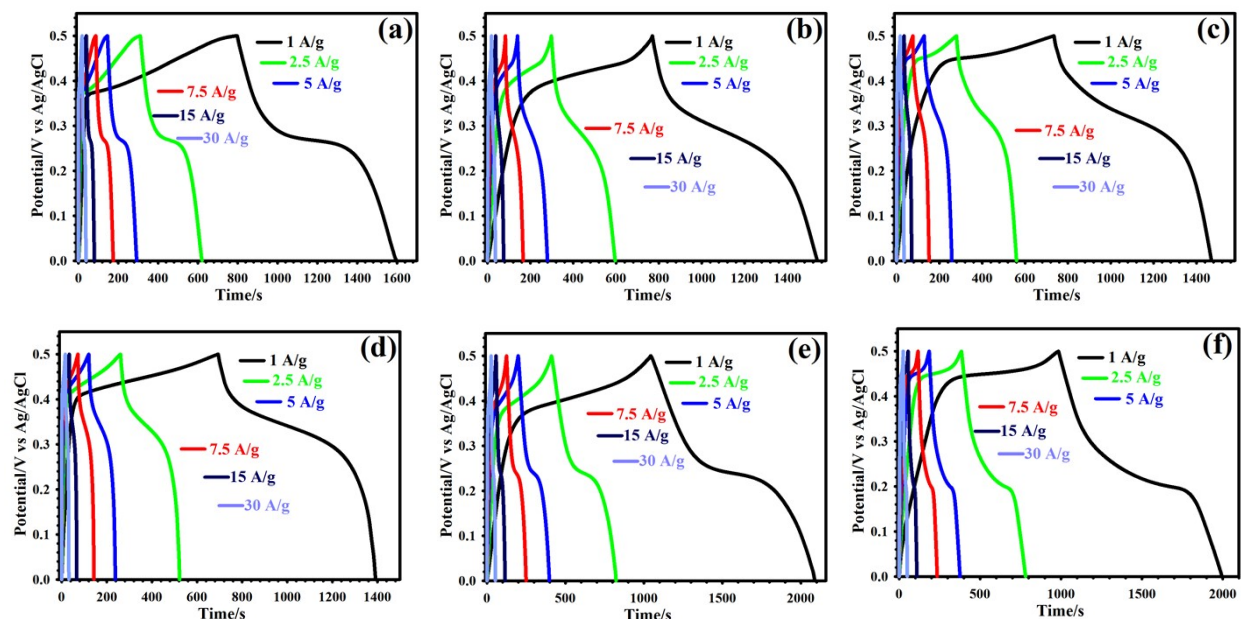


**Fig. 18** (a) CV curves of the NH-CNS-rGO-1, NH-CNS-rGO-2, and NH-CNS-rGO-3 electrodes at 60 mV/s. (b) GCD curves of the NH-CNS-rGO-1, NH-CNS-rGO-2, and NH-CNS-rGO-3 electrodes at 1 A/g. (c) Specific capacities of NH-CNS-rGO-1, NH-CNS-rGO-2, and NH-CNS-rGO-3 electrodes at 1 A/g.

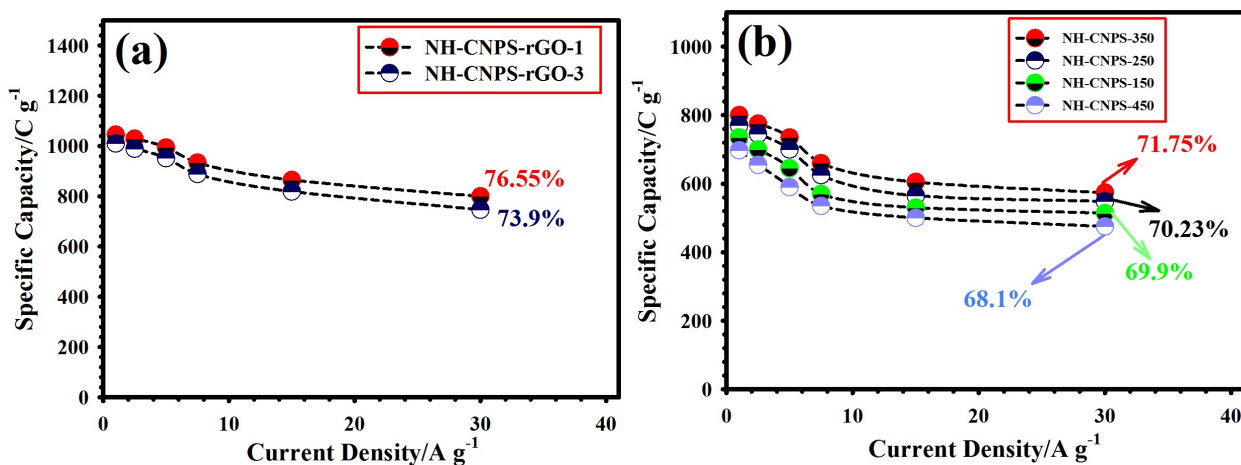




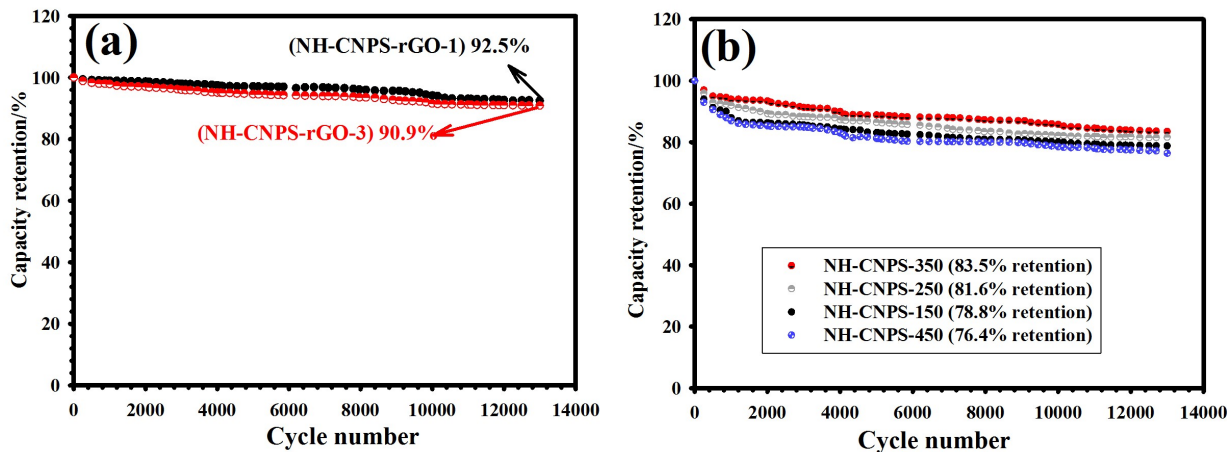
**Fig. S19** (a) CV curves of the NH-CNS-350 electrode at various scan rates from 10 to 100 mV/s. (b) CV curves of the NH-CNS-250 electrode at various scan rates from 10 to 100 mV/s. (c) CV curves of the NH-CNS-150 electrode at various scan rates from 10 to 100 mV/s. (d) CV curves of the NH-CNS-450 electrode at various scan rates from 10 to 100 mV/s. (e) CV curves of the NH-CNS-rGO-1 electrode at various scan rates from 10 to 100 mV/s. (f) CV curves of the NH-CNS-rGO-3 electrode at various scan rates from 10 to 100 mV/s.



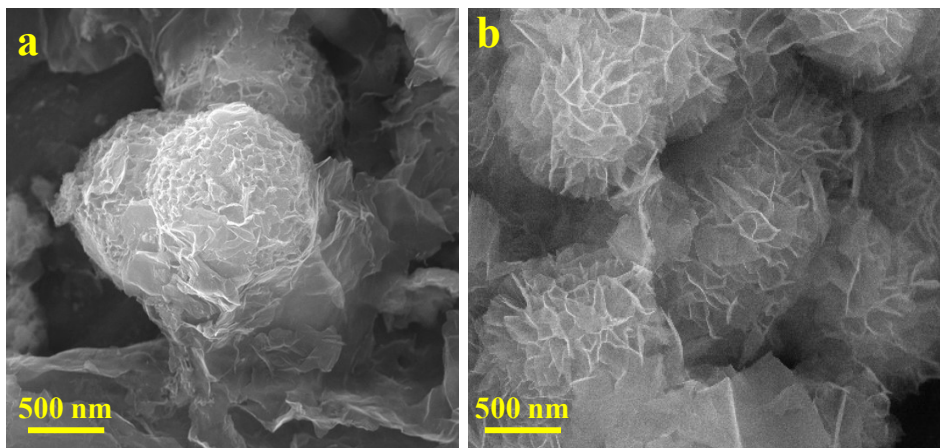
**Fig. S20** (a) GCD curves of the NH-CNS-350 electrode from 1 to 30 A/g. (b) GCD curves of the NH-CNS-250 electrode from 1 to 30 A/g. (c) GCD curves of the NH-CNS-150 electrode from 1 to 30 A/g. (d) GCD curves of the NH-CNS-450 electrode from 1 to 30 A/g. (e) GCD curves of the NH-CNS-rGO-1 electrode from 1 to 30 A/g. (f) GCD curves of the NH-CNS-rGO-3 electrode from 1 to 30 A/g.



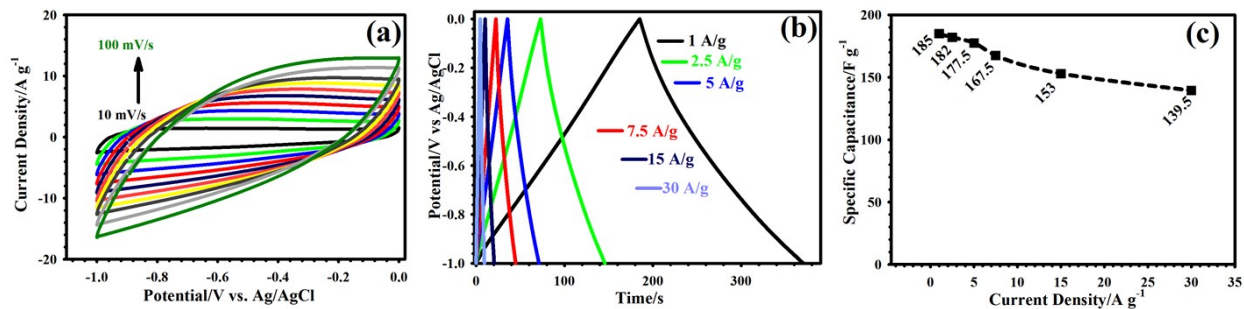
**Fig. S21** (a) Specific capacities vs. current densities of the NH-CNPS-rGO-1 and NH-CNPS-rGO-3. (b) Specific capacities vs. current densities of the NH-CNPS-150, NH-CNPS-250, NH-CNPS-350, and NH-CNPS-450.



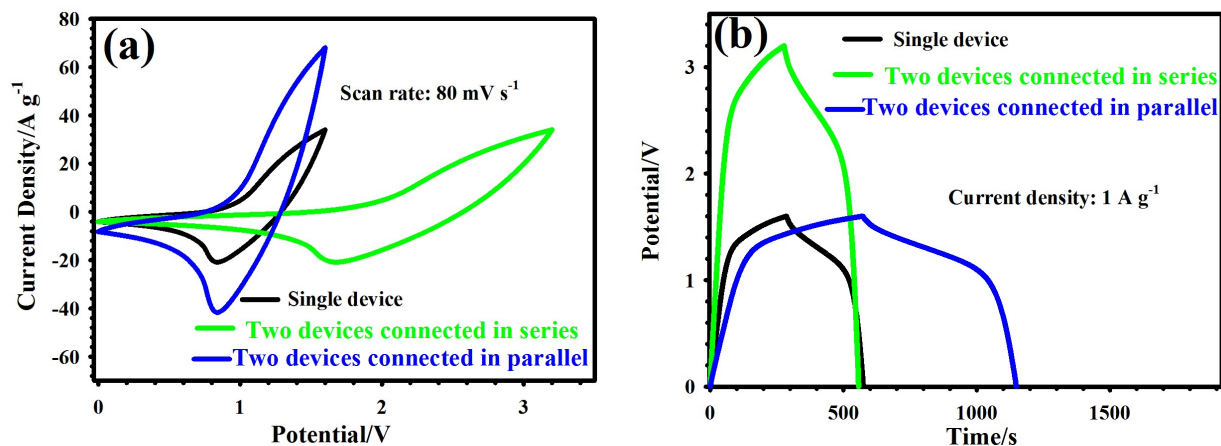
**Fig. S22** (a) Durability of the NH-CNPS-rGO-1 and NH-CNPS-rGO-3 electrodes at 15 A/g. (b) Durability of the NH-CNPS-150, NH-CNPS-250, NH-CNPS-350, and NH-CNPS-450 electrodes at 15 A/g.



**Fig. S23** (a) FE-SEM images of the NH-CNPS-rGO-2 after 13000 GCD cycles. (b) FE-SEM images of the NH-CNPS-350 after 13000 GCD cycles.



**Fig. S24** (a) CVs of the AC electrode from 10 to 100 mV/s. (b) GCD graphs of the AC from 1 to 30 A/g. (c) Specific capacities vs. current densities of the AC electrode.



**Fig. S25** (a) CV curves of single and two devices connected in series and parallel at scan rate of 80 mV/s. (b) GCD curves of single and two devices connected in series and parallel at 1 A g<sup>-1</sup>.

**Table S1.** The as-synthesized precursors and their corresponding synthesis parameters

Entry	Starting materials	Solvothermal Temperature (°C)	Solvothermal Time (h)	mmol ratios of the Cu(CH <sub>3</sub> COO) <sub>2</sub> :Ni(CH <sub>3</sub> COO) <sub>2</sub>	Final precursors
1	CuNi-EG	100	10	3:2	CN-EG32-100
2	CuNi-EG	130	10	3:2	CN-EG32-130
3	CuNi-EG	160	10	3:2	CN-EG32-160
4	CuNi-EG	180	10	3:2	CN-EG32-180 (Optimized sample)

**Table S2.** The as-synthesized precursors and their corresponding synthesis parameters

Entry	Starting materials	Solvothermal Temperature (°C)	Solvothermal Time (h)	mmol ratios of the Cu(CH <sub>3</sub> COO) <sub>2</sub> :Ni(CH <sub>3</sub> COO) <sub>2</sub>	Final precursors
1	CuNi-EG	180	10	1:1	CN-EG11-180
2	CuNi-EG	180	10	1:2	CN-EG12-180
3	CuNi-EG	180	10	2:1	CN-EG21-180
4	CuNi-EG	180	10	2:3	CN-EG23-180
5	CuNi-EG	180	10	3:2	CN-EG32-180 (Optimized sample)

**Table S3.** The as-synthesized phosphide samples and their corresponding synthesis parameters

Entry	Starting materials	Phosphorization Temperature (°C)	Phosphorization Time (h)	Final samples
1	CN-EG180	150	2	NH-CNPS-150
2	CN-EG180	250	2	NH-CNPS-250
3	CN-EG180	350	2	NH-CNPS-350 (Optimized sample)
4	CN-EG180	450	2	NH-CNPS-450

**Table S4.** Comparison of the performance of the NH-CNPS-rGO-2 with other previously reported materials.

Composition	Capacity (C/g)	Cycles, retention	Rate capability	ED (Wh kg <sup>-1</sup> )	Reference
NiCoP	761 at 1 A g <sup>-1</sup>	50000, 90.2%	91.1% at 20 A g <sup>-1</sup>	35.6	1
Ni <sub>2</sub> P/NiCoP	741.3 at 1 A g <sup>-1</sup>	30000, 89.2%	75.5% at 50 A g <sup>-1</sup>	44.5	2
Ni-Co-P/POx/C	583 at 1 A g <sup>-1</sup>	5000, 77.3%	62.7% at 30 A g <sup>-1</sup>	37.59	3
O-Co <sub>x</sub> Ni <sub>y</sub> P	717.1 at 1 A g <sup>-1</sup>	5000, 95.1%	66.7% at 20 A g <sup>-1</sup>	47.5	4
Ni <sub>2</sub> P/Ni/C	257.2 at 1 A g <sup>-1</sup>	5000, 84.3%	60.5% at 10 A g <sup>-1</sup>	25.4	5
Mn-CoP/NF	456 at 0.5 A g <sup>-1</sup>	20000, 89%	77.4% at 10 A g <sup>-1</sup>	14.82	6



Ni <sub>2</sub> P/Co <sub>3</sub> O <sub>4</sub> /N-CQDs	1044 at 1 A g <sup>-1</sup>	6000, 90.5%	83.9% at 20 A g <sup>-1</sup>	53.5	7
PPy@CoP	443 at 1 A g <sup>-1</sup>	5000, 93%	50.5% at 20 A g <sup>-1</sup>	38.1	8
NH-CNPS-rGO-2	1075 at 1 A g <sup>-1</sup>	13000, 94.7%	79.3% at 30 A g <sup>-1</sup>	64	This study

## References

- 1 M. Gao, W.-K. Wang, X. Zhang, J. Jiang and H.-Q. Yu, *J. Phys. Chem. C* 2018, **122**, 25174–25182.
- 2 Z. Li, K. Ma, F. Guo, C. Ji, H. Mi, P. Qiu and H. Pang, *Mater. Lett.* 2021, **288**, 129319.
- 3 X. Zhang, J. Wang, Y. Sui, F. Wei, J. Qi, Q. Meng, Y. He and D. Zhuang, *ACS Appl. Nano Mater.* 2020, **3**, 11945–11954.
- 4 S. Jiang, M. Pang, R. Liu, J. Song, R. Wang, N. Li, Q. Pan, H. Yang, W. He and J. Zhao, *J. Alloys Compd.* 2022, **895**, 162451.
- 5 Y. Xu, S. Xiong, S. Weng, J. Wang, J. Wang, H. Lin, Y. Jiao and J. Chen, *New J. Chem.*, 2020, **44**, 6810-6817.
- 6 M. Zhang, H. Du, Z. Wei, X. Zhang and R. Wang, *ACS Appl. Energy Mater.* 2022, **5**, 186–195
- 7 Z. Ji, K. Liu, L. Chen, Y. Nie, D. Pasang, Q. Yu, X. Shen, K. Xu and S. Premlatha, *J. Colloid Interface Sci.* 2022, **609**, 503-512.
- 8 C. Yue, B. Hu, W. Huang, A. Liu, Z. Guo, J. Mu, X. Zhang, X. Liu and H. Che, *J. Electroanal. Chem.* 2021, **899**, 115656.