

# Polypyrrole nanoparticles embedded nitrogen-doped graphene composites as novel cathode for long life cycles and high-power zinc-ion hybrid supercapacitors

Prasit Pattananuwat,<sup>† a,b,c</sup> Rojana Pornprasertsuk,<sup>a,b,c</sup> Jiaqian Qin<sup>b,d</sup> and Suchitra Prasertkaew<sup>e</sup>

<sup>a</sup>*Department of Materials Science, Faculty of Science, Chulalongkorn University, Bangkok, 10330, Thailand.*

<sup>b</sup>*Research Unit of Advanced Materials for Energy Storage, Chulalongkorn University, Bangkok, Thailand*

<sup>c</sup>*Center of Excellence on Petrochemical and Materials Technology, Chulalongkorn University, Bangkok, Thailand*

<sup>d</sup>*Metallurgy and Materials Science Research Institute, Chulalongkorn University, Bangkok 10330, Thailand*

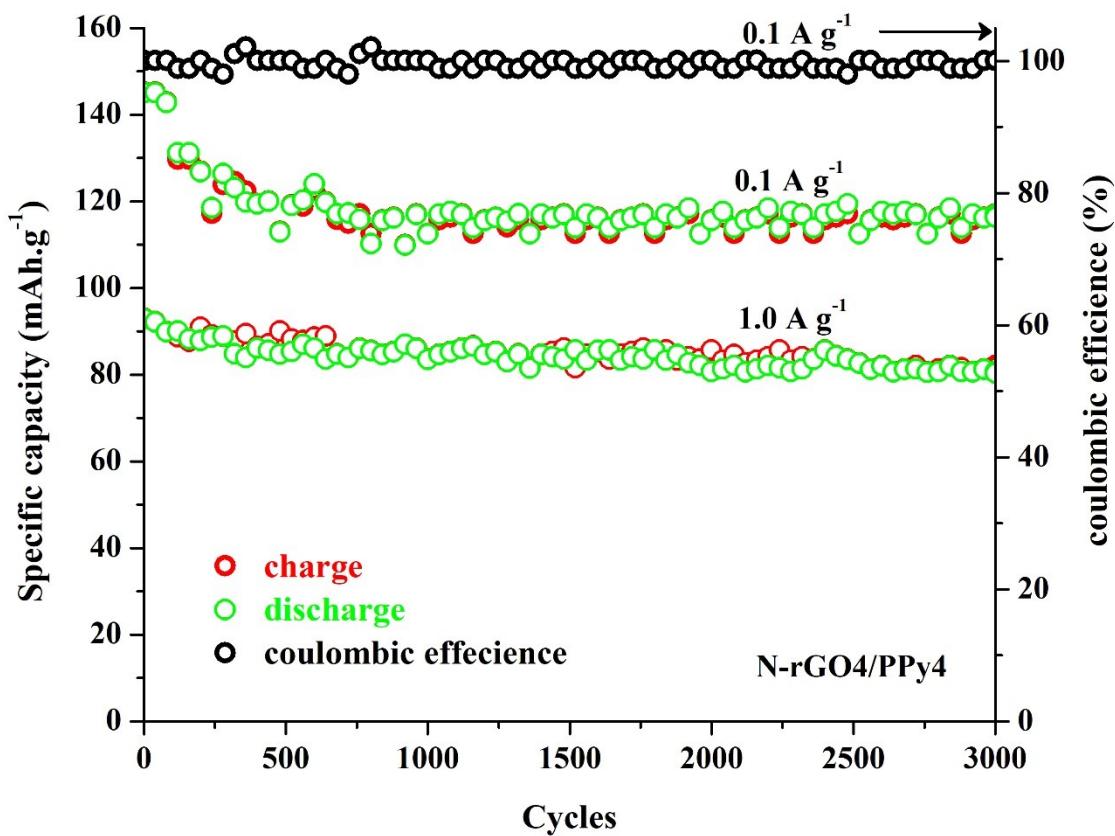
<sup>e</sup>*Petrochemistry and Polymer Science, Faculty of Science, Chulalongkorn University, Bangkok, Thailand*

<sup>†</sup> *These are corresponding authors*

*Email:* [prasit.pat@chula.ac.th](mailto:prasit.pat@chula.ac.th)

**Table S1** the lists of ZICs/ZHSCs reports

Materials	Electrolyte	Specific capacitance	Specific capacity	Energy/Power density	Retention	Ref.
Hollow carbon spheres	ZnSO <sub>4</sub> PAM	-	86.8 mA h g <sup>-1</sup> at 0.5 A g <sup>-1</sup>	59.7 Wh kg <sup>-1</sup> at 447.8 W kg <sup>-1</sup>	98% after 15000 cycles at 1.0 A g <sup>-1</sup>	1
Coconut shells derived activated carbon	1.0 M Zn(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub> in AN	170 F g <sup>-1</sup> at 0.1 A g <sup>-1</sup>	-	52.7 Wh kg <sup>-1</sup> at 1725 W kg <sup>-1</sup>	85% after 22000 cycles at 2.0 A g <sup>-1</sup>	2
Kelp-Carbon	Zn(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub> PAM	445 F g <sup>-1</sup> at 0.1 A g <sup>-1</sup>	196.7 mAh g <sup>-1</sup> at 0.1 A g <sup>-1</sup>	111.5 Wh kg <sup>-1</sup> at 1300 W kg <sup>-1</sup>	88% after 4000 cycles at 0.1 A g <sup>-1</sup>	3
Activated graphene	3.0 M Zn(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub>	210 F g <sup>-1</sup> at 0.1 A g <sup>-1</sup>	-	31.4 kW kg <sup>-1</sup> at 106.3 Wh kg <sup>-1</sup>	93% after 80000 cycles at 8.0 A g <sup>-1</sup>	4
ZnMn <sub>2</sub> O <sub>4</sub> /carbon	3.0 M Zn(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub>	-	120 mAh g <sup>-1</sup> at 0.05 A g <sup>-1</sup>	~202 Wh kg <sup>-1</sup>	94% after 500 cycles at 0.5 A g <sup>-1</sup>	5
Nanostructured PPy composite	2.0 M ZnCl <sub>2</sub> +3.0 M NH <sub>4</sub> Cl	-	151.1 mAh g <sup>-1</sup> at 0.05 A g <sup>-1</sup> , 125 mAh g <sup>-1</sup> at 0.1 A g <sup>-1</sup>	11.7 kW kg <sup>-1</sup> at 64.0 Wh kg <sup>-1</sup>	76.7% after 1000 cycles at 8.0 A g <sup>-1</sup>	6
Hollow mesoporous-carbon nanospheres	2.0 M ZnSO <sub>4</sub> +1.0 M Na <sub>2</sub> SO <sub>4</sub>	212.1 F g <sup>-1</sup> at 0.2 A g <sup>-1</sup>	-	75.4 Wh kg <sup>-1</sup> at 0.16 kW kg <sup>-1</sup>	99.4% after 2500 cycles at 2.0 A g <sup>-1</sup>	7
AC raw material	2.0 M ZnSO <sub>4</sub>	-	121 mAh g <sup>-1</sup>	84 Wh kg <sup>-1</sup> at 14.9 kW kg <sup>-1</sup>	91% after 10000 cycles at 1.0 A g <sup>-1</sup>	8
Bamboo-derived porous carbons	2.0 M ZnSO <sub>4</sub>	-	51.4 mA h g <sup>-1</sup>	12.1 Wh kg <sup>-1</sup> at 993.4 W kg <sup>-1</sup>	99 % after 2500 cycles at 1.0 A g <sup>-1</sup>	9
Mesoporous structured activated carbon	2.0 M ZnSO <sub>4</sub> (two electrode testing)	-	176 mAh g <sup>-1</sup> at 0.5 A g <sup>-1</sup> , 80 mAh g <sup>-1</sup> at 7.0 A g <sup>-1</sup>	188 Wh kg <sup>-1</sup> at 533 W kg <sup>-1</sup>	78% after 40000 cycles 10.0 A g <sup>-1</sup>	10
Porous carbon nanosheets	ZnCl <sub>2</sub> deep-eutectic-solvent electrolyte	-	78.6 mAh g <sup>-1</sup> at 0.5 A g <sup>-1</sup>	52.8 W h kg <sup>-1</sup> at 384.8 W kg <sup>-1</sup>	-	11
Three-dimensional (3D) graphene	1.0 M ZnSO <sub>4</sub> electrolyte	222.03 F g <sup>-1</sup> at 0.5 A g <sup>-1</sup>	-	118.42 Wh L <sup>-1</sup> at 24.00 kW L <sup>-1</sup>	80% after 30000 cycles at 10 A g <sup>-1</sup>	12
Hierarchically porous carbon	3.0 M Zn(CF <sub>3</sub> SO <sub>3</sub> ) <sub>2</sub>	44.3 F g <sup>-1</sup> at 0.5 A g <sup>-1</sup>	-	6.2 W h kg <sup>-1</sup> at 250 W kg <sup>-1</sup>	> 98% after 10000 cycles at 2.0 A g <sup>-1</sup>	13
ZN@graphene /AC cathodes	2.0 M ZnSO <sub>4</sub> electrolyte	186 F g <sup>-1</sup> at 0.1 A g <sup>-1</sup>	-	78.32-30.26 Wh kg <sup>-1</sup> at 30.26- 8010 W kg <sup>-1</sup>	> 80 % after 10000 cycles 0.1 A g <sup>-1</sup>	14
3D MXene (Ti <sub>3</sub> C <sub>2</sub> Tx)-reduced graphene oxide	ZnSO <sub>4</sub>	128.6 F g <sup>-1</sup> 0.4 A g <sup>-1</sup>	-	34.9 Wh kg <sup>-1</sup> at 279.9 W kg <sup>-1</sup>	95 % after 75000 at 5.0 A g <sup>-1</sup>	15
Oxygen-enriched porous carbon	gelatin/ZnSO <sub>4</sub>	-	132.7 mAh g <sup>-1</sup> at 0.2 A g <sup>-1</sup>	38.6 W h kg <sup>-1</sup> at 3760 W kg <sup>-1</sup>	-	16
N-rGO/PPy	2.0 M ZnSO <sub>4</sub>	385 F g <sup>-1</sup> at 0.1 A g <sup>-1</sup>	145.32 mA h g <sup>-1</sup> at 0.1 A g <sup>-1</sup> , 69 mAh g <sup>-1</sup> at 7.0 A g <sup>-1</sup>	232.50 - 110.40 W h k g <sup>-1</sup> at 160 - 11200 W k g <sup>-1</sup>	82.5% after 10000 cycles at 7.0 A g <sup>-1</sup>	(This work)



**Fig. S1** The rate capacity of N-rGO4/PPy4 at 0.1 and 1.0 A g<sup>-1</sup> for 3000 cycles

## **References**

1. S. Chen, L. Ma, K. Zhang, M. Kamruzzaman, C. Zhi and J. A. Zapien, *Journal of Materials Chemistry A*, 2019, **7**, 7784-7790.
2. H. Wang, M. Wang and Y. Tang, *Energy Storage Materials*, 2018, **13**, 1-7.
3. J. Zeng, L. Dong, L. Sun, W. Wang, Y. Zhou, L. Wei and X. Guo, *Nano-Micro Letters*, 2020, **13**, 19.
4. S. Wu, Y. Chen, T. Jiao, J. Zhou, J. Cheng, B. Liu, S. Yang, K. Zhang and W. Zhang, *Advanced Energy Materials*, 2019, **9**, 1902915.
5. N. Zhang, F. Cheng, Y. Liu, Q. Zhao, K. Lei, C. Chen, X. Liu and J. Chen, *Journal of the American Chemical Society*, 2016, **138**, 12894-12901.
6. X. Li, X. Xie, R. Lv, B. Na, B. Wang and Y. He, *Energy Technology*, 2019, **7**, 1801092.
7. S. Chen, G. Yang, X. Zhao, N. Wang, T. Luo, X. Chen, T. Wu, S. Jiang, P. A. van Aken, S. Qu, T. Li, L. Du, J. Zhang, H. Wang and H. Wang, *Frontiers in Chemistry*, 2020, **8**, 663.
8. L. Dong, X. Ma, Y. Li, L. Zhao, W. Liu, J. Cheng, C. Xu, B. Li, Q.-H. Yang and F. Kang, *Energy Storage Materials*, 2018, **13**, 96-102.
9. H. Chen, Y. Zheng, X. Zhu, W. Hong, Y. Tong, Y. Lu, G. Pei, Y. Pang, Z. Shen and C. Guan, *Materials Research Bulletin*, 2021, **139**, 111281.
10. G.-H. An, *Applied Surface Science*, 2020, **530**, 147220.
11. H. Tian, R. Cheng, L. Zhang, Q. Fang, P. Ma, Y. Lv and F. Wei, *Materials Letters*, 2021, **301**, 130237.
12. L. Zhang, D. Wu, G. Wang, Y. Xu, H. Li and X. Yan, *Chinese Chemical Letters*, 2021, **32**, 926-931.
13. H. Li, J. Wu, L. Wang, Q. Liao, X. Niu, D. Zhang and K. Wang, *Chemical Engineering Journal*, 2022, **428**, 131071.
14. X. Zhang, C. Chen, S. Gao, X. Luo, Y. Mo, B. Cao and Y. Chen, *Journal of Energy Storage*, 2021, **42**, 103037.
15. Q. Wang, S. Wang, X. Guo, L. Ruan, N. Wei, Y. Ma, J. Li, M. Wang, W. Li and W. Zeng, *Advanced Electronic Materials*, 2019, **5**, 1900537.
16. Y. Zheng, W. Zhao, D. Jia, Y. Liu, L. Cui, D. Wei, R. Zheng and J. Liu, *Chemical Engineering Journal*, 2020, **387**, 124161.