# **Fabrication of Fe3O4-incorporated MnO<sup>2</sup> nanoflowers as electrodes for**

# **enhanced asymmetric supercapacitor performance**

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### **Figure Content**

- **Figure S1.** N<sub>2</sub> ads/des isotherms of the pristine Fe<sub>3</sub>O<sub>4</sub>, pristine MnO<sub>2</sub> and MnO<sub>2</sub>@Fe<sub>3</sub>O<sub>4</sub> nanoflower like structure.
- **Figure S2.** BJH curves of the pristine  $Fe<sub>3</sub>O<sub>4</sub>$ , pristine  $MnO<sub>2</sub>$  and  $MnO<sub>2</sub>(QFe<sub>3</sub>O<sub>4</sub>$  nanoflower like structure.
- **Figure S3.** Nyquist plot of the Fe<sub>3</sub>O<sub>4</sub>, MnO<sub>2</sub> and MnO<sub>2</sub>@Fe<sub>3</sub>O<sub>4</sub> nanoflowers.
- **Figure S4.** CV for the activated carbon for the  $Fe<sub>3</sub>O<sub>4</sub>$  at various scan rates in the potential range from -1.0 V to 0 V.

## **S5. Calculations for the loading mass**

**Table S1.** Comparison of specific capacitance of MnO<sub>2</sub>@Fe<sub>3</sub>O<sub>4</sub> nanoflowers reported to date with those in the present study.

## **References**



**Figure S1.** N<sub>2</sub> ads/des isotherms of the pristine Fe<sub>3</sub>O<sub>4</sub>, pristine MnO<sub>2</sub> and MnO<sub>2</sub>@Fe<sub>3</sub>O<sub>4</sub> nanoflower like structure.



**Figure S2.** BJH curves of the pristine Fe<sub>3</sub>O<sub>4</sub>, pristine  $MnO_2$  and  $MnO_2@Fe_3O_4$  nanoflower like structure.



**Figure S3.** Nyquist plot of the pristine Fe<sub>3</sub>O<sub>4</sub>, pristine MnO<sub>2</sub> and MnO<sub>2</sub>@Fe<sub>3</sub>O<sub>4</sub> nanoflowers.



**Figure S4.** CV for the activated carbon at various scan rates in the potential range from -1.0 V to

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0 V.

#### **S5. Calculations for the loading mass**

The mass ratio was calculated from the following equations to obtain charge balance

$$
\frac{m_+}{m_-} = \frac{C_{s-} \times \Delta V}{C_{s+} \times \Delta V} \tag{1}
$$

where  $m<sup>+</sup>$  and  $m<sup>-</sup>$  are masses on positive (cathode) and negative (anode) electrodes of the ASC, Cs+ and Cs− are specific capacitances of the electrode materials in the potential windows of ΔVand  $\Delta V$ +, respectively, measured against a reference electrode in the three electrode configurations. The mass loading ratio of the positive and negative electrodes was estimated as ∼5.7, according to eq 1. Approximately 1.5 mg of negative electrode (AC) and 0.9 mg of positive electrode  $(MnO<sub>2</sub>(QFe<sub>3</sub>O<sub>4</sub>)$  nanoflower) were used to maintain the charge neutrality of ASC. Electrochemical assessment of ASC was carried out on the basis of total mass loading on both electrodes. In this study, both electrodes were obtained using the three-electrode system over a potential window of 0 to 0.6 V for the  $MnO<sub>2</sub>(QFe<sub>3</sub>O<sub>4</sub>$  nanoflower cathode and 0 to -1.0 V for the AC anode electrode, which collectively provide a large potential window of upto 1.6 V for the  $MnO<sub>2</sub>(QFe<sub>3</sub>O<sub>4</sub>)/AC$  nanoflower ASC device. Therefore, the initial voltage is 0 and final provided voltage is 1.6. So,  $\Delta V = 1.6 - 0 = 1.6$  V, which is fixed for our study.

Table S1. Comparison of specific capacitance of MnO<sub>2</sub>@Fe<sub>3</sub>O<sub>4</sub> nanoflowers reported to date with those in the present study.



### **Reference:**

- [1] I. Rabani, J. Yoo, H.-S. Kim, S. Hussain, K. Karuppasamy, and Y.-S. Seo, "Highly dispersive Co 3 O 4 nanoparticles incorporated into a cellulose nanofiber for a highperformance flexible supercapacitor," *Nanoscale,* vol. 13, no. 1, pp. 355-370, 2021.
- [2] Z. Zhu, Z. Wang, Z. Yan, R. Zhou, Z. Wang, and C. Chen, "Facile synthesis of MOFderived porous spinel zinc manganese oxide/carbon nanorods hybrid materials for supercapacitor application," *Ceramics International,* vol. 44, no. 16, pp. 20163-20169, 2018.
- [3] C. J. Raj *et al.*, "Two-dimensional planar supercapacitor based on zinc oxide/manganese oxide core/shell nano-architecture," *Electrochimica Acta,* vol. 247, pp. 949-957, 2017.
- [4] S. Li *et al.*, "Three-dimensional MnO2 nanowire/ZnO nanorod arrays hybrid nanostructure for high-performance and flexible supercapacitor electrode," *Journal of Power Sources,* vol. 256, pp. 206-211, 2014.
- [5] C.-K. Sim, S. R. Majid, and N. Z. Mahmood, "Durable porous carbon/ZnMn2O4 composite electrode material for supercapacitor," *Journal of Alloys and Compounds,* vol. 803, pp. 424-433, 2019.
- [6] X. He, J. E. Yoo, M. H. Lee, and J. Bae, "Morphology engineering of ZnO nanostructures for high performance supercapacitors: enhanced electrochemistry of ZnO nanocones compared to ZnO nanowires," *Nanotechnology,* vol. 28, no. 24, p. 245402, 2017.
- [7] G. Sun, L. Ma, J. Ran, X. Shen, and H. Tong, "Incorporation of homogeneous Co 3 O 4 into a nitrogen-doped carbon aerogel via a facile in situ synthesis method: implications for high

performance asymmetric supercapacitors," *Journal of Materials Chemistry A,* vol. 4, no. 24, pp. 9542-9554, 2016.

- [8] W. Li *et al.*, "Urchin-like MnO2 capped ZnO nanorods as high-rate and high-stability pseudocapacitor electrodes," *Electrochimica Acta,* vol. 186, pp. 1-6, 2015.
- [9] E. Samuel, B. Joshi, Y.-i. Kim, A. Aldalbahi, M. Rahaman, and S. S. Yoon, "ZnO/MnO x nanoflowers for high-performance supercapacitor electrodes," *ACS Sustainable Chemistry & Engineering,* vol. 8, no. 9, pp. 3697-3708, 2020.
- [10] Y. Zhao, P. Jiang, and S.-S. Xie, "ZnO-template-mediated synthesis of three-dimensional coral-like MnO2 nanostructure for supercapacitors," *Journal of power sources,* vol. 239, pp. 393-398, 2013.
- [11] M. Huang *et al.*, "Hierarchical ZnO@ MnO2 core-shell pillar arrays on Ni foam for binderfree supercapacitor electrodes," *Electrochimica Acta,* vol. 152, pp. 172-177, 2015.
- [12] Y. Wang *et al.*, "Ni–Co Selenide Nanosheet/3D Graphene/Nickel Foam Binder-Free Electrode for High-Performance Supercapacitor," *ACS applied materials & interfaces,* vol. 11, no. 8, pp. 7946-7953, 2019.
- [13] H. Fu *et al.*, "Designed formation of NiCo2O4 with different morphologies self-assembled from nanoparticles for asymmetric supercapacitors and electrocatalysts for oxygen evolution reaction," *Electrochimica Acta,* vol. 296, pp. 719-729, 2019.