## **Electronic Supplementary Information**

## Synthesis of Flower-like Manganese Oxide for Accelerated Surface Redox Reactions on Nitrogen-Rich Graphene for Sustainable Aqueous Energy Storage

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Figure S1. SEM images of MF, M@G, and MF@NG



Figure S2. SEM images of a) GO and b) NGO

|    | C (%) | H (%) | N (%) | S (%) | O (%) |
|----|-------|-------|-------|-------|-------|
| GO | 36.12 | 2.61  | 0.00  | 4.85  | 56.42 |
| NG | 66.14 | 1.66  | 10.36 | 0.04  | 21.80 |

 Table S1.
 Elements Analysis of GO and NGO.



Figure S3. a) Raman spectroscopy of GO and NGO. b) XPS survey scan of GO and NGO.



Figure S4. a) Pore distribution of MF, M@G, and MF@NG. b) XPS survey scan of MF@NG.

|   | MF    | M@G    | MF@NG  |
|---|-------|--------|--------|
| Specific surface area (m <sup>2</sup> g <sup>-1</sup> ) | 75.37 | 153.18 | 190.95 |
| Pore volume (cm <sup>3</sup> g <sup>-1</sup> )          | 0.32  | 0.32   | 0.40   |
| Average pore diameter (nm)                              | 3.42  | 3.85   | 3.84   |

**Table S2.** Specific surface area, pore volume, and average pore diameter of MF, M@G, and MF@NG.



**Figure S5.** TEM image of MF@NG. a) High magnification TEM image of MF@NG sample with Mn<sub>3</sub>O<sub>4</sub> part, and a-inset) FFT pattern of corresponding planes. a-i) Measuring d-spacing and corresponding profile plot of the calibration. b) High magnification TEM image of MF@NG sample with GO part, and b-inset) FFT pattern of corresponding plane. b-i) Measuring d-spacing and corresponding profile plot of the calibration.



**Figure S6.** Electrochemical performance of MF electrode. a) CV curves with various scan rates of 5~50 mV s<sup>-1</sup>. b) GCD curves at various specific current of 0.1–2 A g<sup>-1</sup>.



**Figure S7.** Electrochemical performance of M@G electrode. a) CV curves with various scan rates of 5~50 mV s<sup>-1</sup>. b) GCD curves at various specific current of 0.1–2 A g<sup>-1</sup>. c) Diffusion-, and capacitive-contribution reinterpreted from CV curves.

| Materials  | Voltage<br>range (V) | Electrolyte                                  | Specific<br>capacitance (F g <sup>-1</sup> ) | References  |
|--|----------------------|--|--|-------------|
| Bowl-like MnO <sub>2</sub><br>nanosheets                                 | 0-0.8                | 1M Na <sub>2</sub> SO <sub>4</sub>           | 379  | [23]        |
| MnO <sub>2</sub> nanorods electrode                                      | 0–0.8                | 0.5M Na <sub>2</sub> SO <sub>4</sub>         | 485  | [24]        |
| MnO <sub>2</sub> nanosheets  | 0-1.0                | SiO <sub>2</sub> -LiTFSI gel                 | 243  | [25]        |
| Nitrogen doped<br>graphene<br>foam/carbon<br>nanotube/manganese<br>oxide | -0.2–0.8             | 1M Na <sub>2</sub> SO <sub>4</sub>           | 284  | [26]        |
| MnO <sub>2</sub> /carbon<br>nanotube/polyimide                           | 0-1.1                | CMC/Na <sub>2</sub> SO <sub>4</sub> gel      | 543  | [27]        |
| Manganese<br>oxide/carbon aerogel  | 0.1–0.9              | $0.5 \mathrm{M} \mathrm{Na}_2 \mathrm{SO}_4$ | 503  | [28]        |
| Reduced graphene<br>oxide/MnO <sub>2</sub>                               | 0–1.0                | 1M Na <sub>2</sub> SO <sub>4</sub>           | 188  | [29]        |
| Mn <sub>2</sub> O <sub>3</sub> -carbon                                   | -0.3-0.3             | 6M KOH                                       | 600  | [30]        |
| Porous MnO <sub>2</sub><br>nanosheets                                    | 0–0.9                | 0.5M MgSO <sub>4</sub>                       | 329  | [31]        |
| Potassium-containing<br>manganese oxide                                  | 0-1.0                | 0.1M Na <sub>2</sub> SO <sub>4</sub>         | 234  | [32]        |
| Nitrogen doped<br>graphene/Mn <sub>3</sub> O <sub>4</sub>                | 0–0.8                | 1M Na <sub>2</sub> SO <sub>4</sub>           | 654  | [This work] |

Table S3. Comparison of the electrochemical performance of the various Mn-oxide based materials

![](_page_11_Figure_0.jpeg)

**igure S8.** Electrochemical performance of AC electrode. a) CV curves with various scan rates of 5-50 mV s<sup>-1</sup>. b) ElS profile at frequency of 1 M-0.01 Hz with 5mV amplitude. c) GCD curves at various specific current of 0.1-2 A g<sup>-1</sup>. d) Gravimetric capacitance reinterpreted from GCD curves.