

## Supporting Information

### Ranunculus-like Core-shell Ni(OH)<sub>2</sub>@Mn<sub>2</sub>O<sub>3</sub> as a High Specific Capacitance Cathode Material for Alkaline Supercapacitors

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#### Experimental

##### Material synthesis:

All experiments used analytic grade reagents and ultrapure water. Ranunculus flower-like Ni(OH)<sub>2</sub>@Mn<sub>2</sub>O<sub>3</sub> with core-shell structure were prepared by a two-step procedure. Firstly, ranunculus flower-like Ni(OH)<sub>2</sub> was synthesized using 2 mmol of Ni(CH<sub>3</sub>COO)<sub>2</sub> dissolved in 57.5 mL deionized water, to which 2.5 mL of glycerol was added drop-wise under magnetic stirring to form a homogeneous blue solution. The solution was transferred to a 100 mL Teflon-lined stainless steel autoclave, which was then sealed and oven heated at 200 °C for 6 h. Upon unassisted cooling to room temperature, the resultant greenish precipitate was then separated by centrifugation with repeated washing with distilled water and ethanol, and then finally dried at 60 °C in air.

Ni(OH)<sub>2</sub>@Mn<sub>2</sub>O<sub>3</sub> was prepared by dispersing the as-prepared Ni(OH)<sub>2</sub> in 5 mL deionized water, followed by addition of different amounts of KMnO<sub>4</sub> (0.002, 0.01, 0.02, 0.05, and 0.1 mol L<sup>-1</sup>) solution under magnetic stirring at room temperature. The reactions were held for 10 min, and the light-brown Ni(OH)<sub>2</sub>@Mn<sub>2</sub>O<sub>3</sub> resultant product was centrifuged, washed with distilled water and ethanol, and finally dried at 60 °C in air.

##### Characterization

XRD patterns were recorded on a Shimadzu XD-3A (Japan) using filtered Cu-Kα radiation (λ = 0.15418 nm)

generated at 40 kV and 30 mA. Scans for  $2\theta$  values were recorded at  $4^\circ \text{ min}^{-1}$ . Scanning electron microscopy (SEM) images were obtained using a Carl Zeiss Ultra Plus. Transmission electron microscopy (TEM) high angle annular dark field scanning transmission electron microscopy (STEM) images of the catalysts were obtained using a JEOL (JEM-2000 FX) microscope operating at 200 kV. Raman spectroscopy was recorded on Bruker Senterra Raman spectrometer with 532 nm wavelength excitation at 2 mW power. Specific surface area was determined by Brunauer-Emmett-Teller (BET) method, and the BJH method was used for analyzing the full range of pore size distribution based on the desorption isotherms obtained on a Quantachrome Autosorb-1 volumetric analyzer. X-Ray Photoelectron Spectroscopy (XPS) spectra were generated using a PHI-5702 multifunctional X-ray photoelectron spectrometer (American). Binding energies were determined by referencing to the C 1 s peak at 285.0 eV.

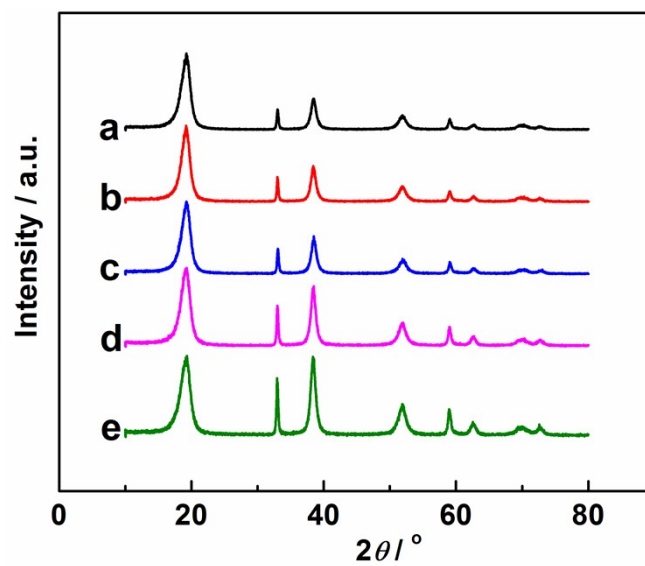
### **Electrochemical measurements**

Cyclic voltammograms (CV) and galvanostatic charge/discharge tests on  $\text{Ni}(\text{OH})_2@\text{Mn}_2\text{O}_3$  were carried out in a three-electrode cell system. The working electrode comprised a film of  $\text{Ni}(\text{OH})_2@\text{Mn}_2\text{O}_3$ , carbon black and poly(tetrafluoroethylene) with a mass ratio of 80:10:10, pressed into Ni foam current collectors. An Hg/HgO (1.0 M KOH) reference electrode and activated carbon counter electrode in  $6 \text{ mol L}^{-1} \text{ KOH} + 0.3 \text{ mol L}^{-1} \text{ LiOH}$  electrolyte was used in all experiments. CV tests were done on a CHI 650D electrochemical workstation. Galvanostatic charge/discharge tests were carried out on a Neware Battery Tester (BTS6.0, Neware Technology Company, Guangdong, China).

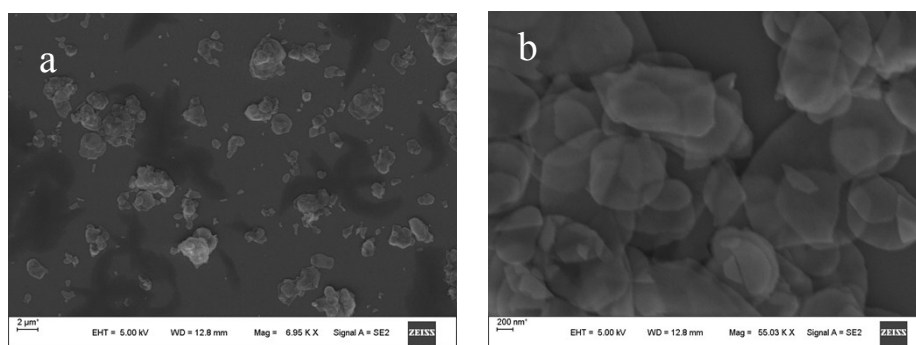
The capacitance of the electrode (C) was calculated according to the following equation based on CVs or the discharge curves.

$$C = \frac{Q}{V} = \frac{i\Delta t}{\Delta V m} \quad (1)$$

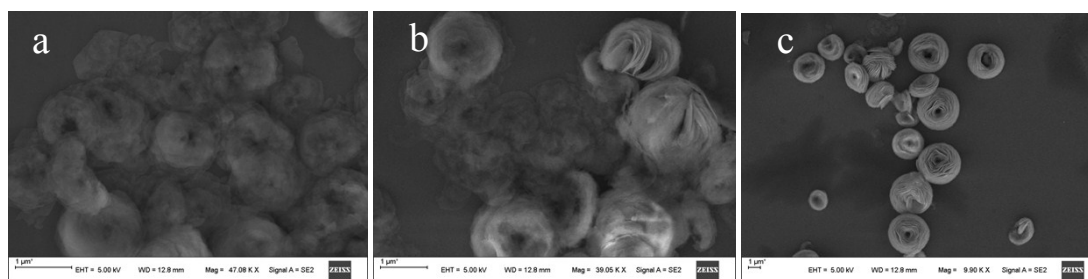
where  $i$  is the sampled current,  $\Delta t$  is a sampling time span,  $\Delta V$  is the total potential deviation of the voltage window, and  $m$  is the mass of the active material examined.



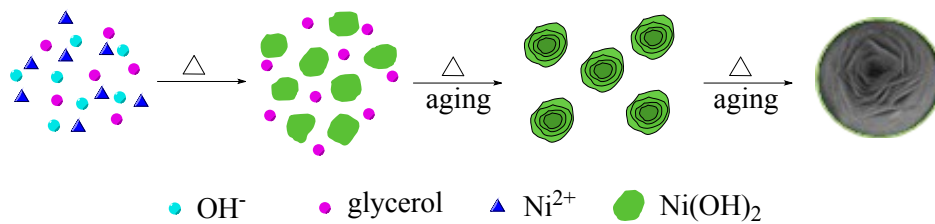
**Fig. S1** XRD patterns of  $\text{Ni}(\text{OH})_2@\text{Mn}_2\text{O}_3$  prepared using different  $\text{KMnO}_4$  concentrations “a-e” at 0.002, 0.01, 0.02, 0.05 and 0.1 mol  $\text{L}^{-1}$  respectively.



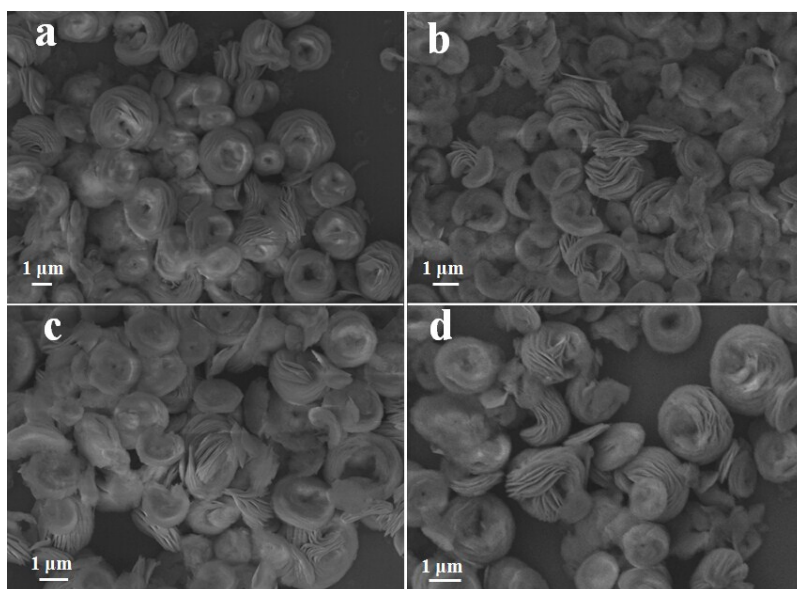
**Fig. S2** SEM images of  $\text{Ni}(\text{OH})_2@\text{Mn}_2\text{O}_3$  prepared in the absence of glycerol.



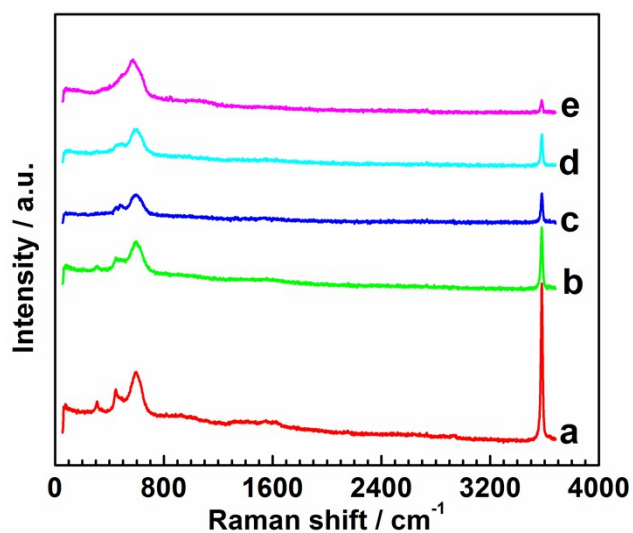
**Fig. S3** SEM images of  $\text{Ni}(\text{OH})_2@\text{Mn}_2\text{O}_3$  prepared with 30 min (a), 1 h (b) and 6 h (c) of the reaction time.



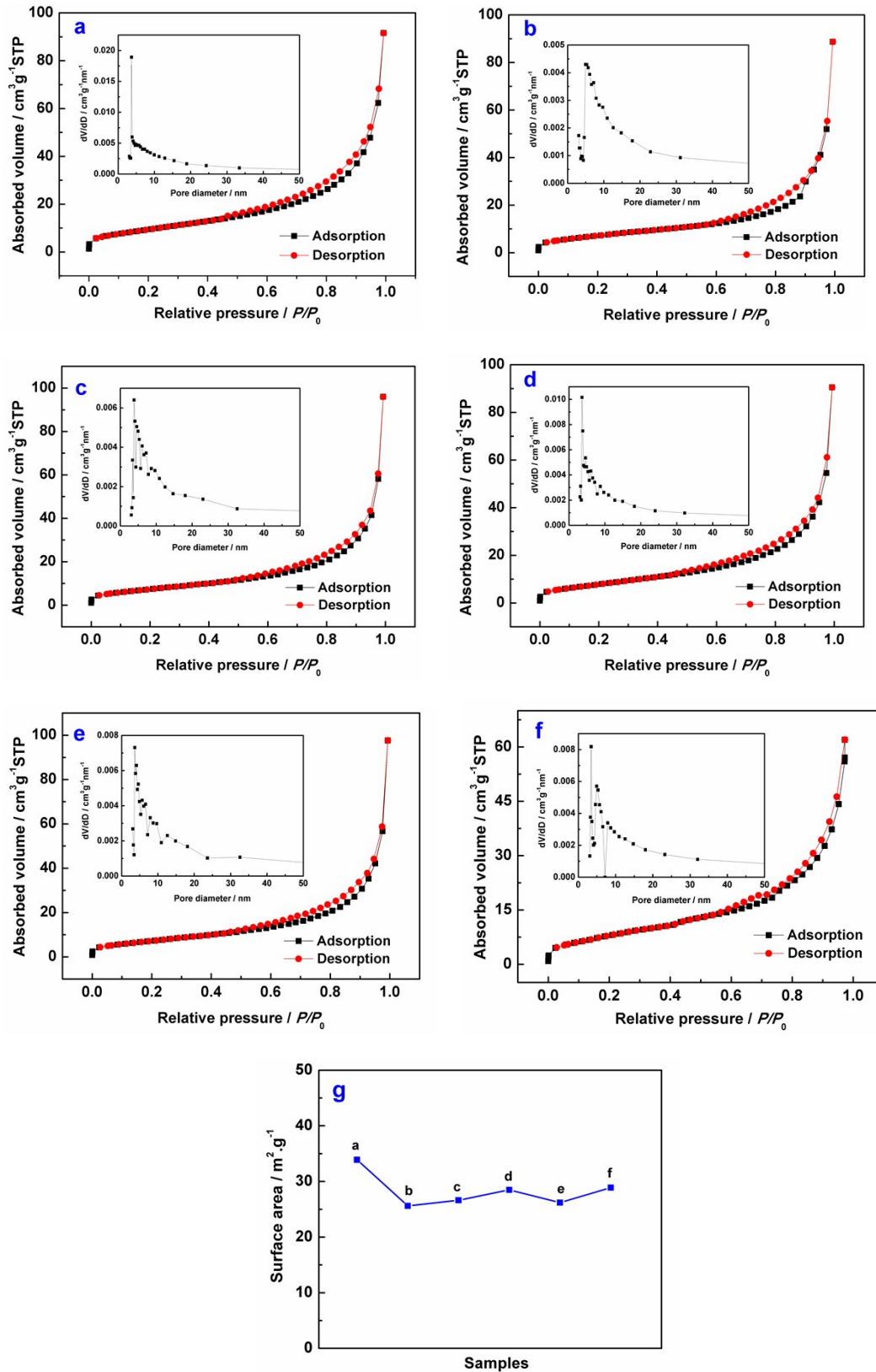
**Fig. S4** Mechanism for the formation of ranunculus flower-like Ni(OH)<sub>2</sub>@Mn<sub>2</sub>O<sub>3</sub>.



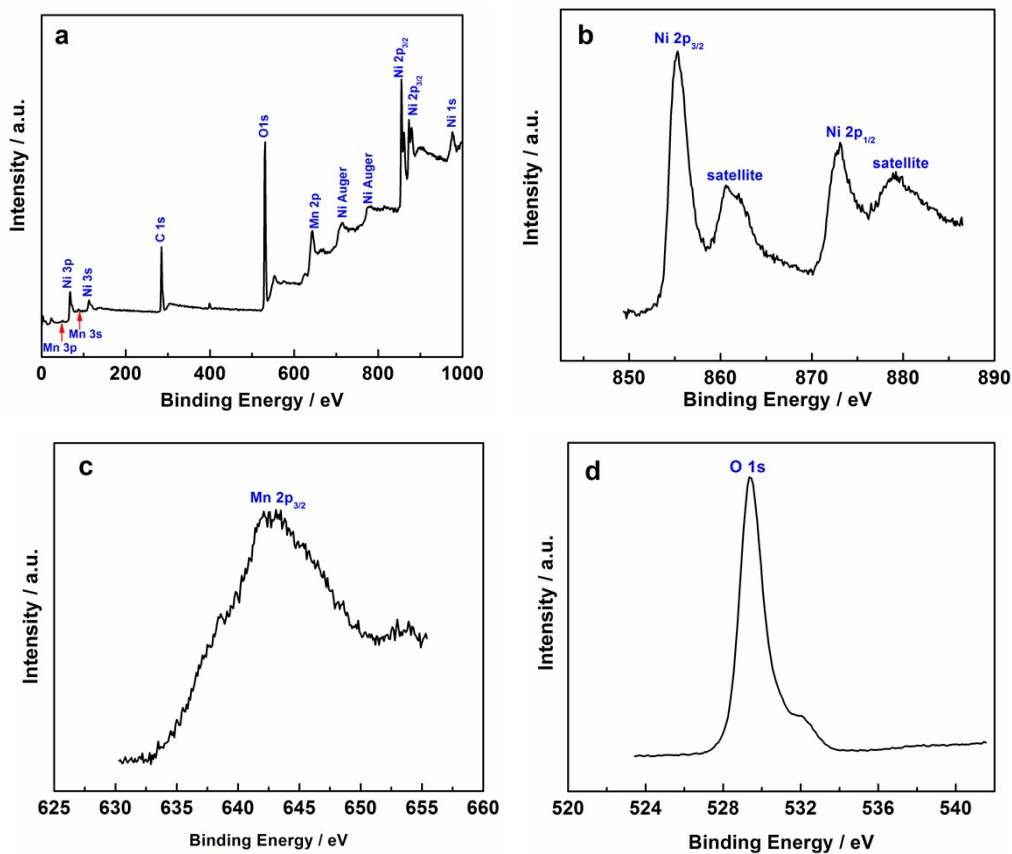
**Fig. S5** SEM images of Ni(OH)<sub>2</sub>@Mn<sub>2</sub>O<sub>3</sub> prepared using KMnO<sub>4</sub> concentrations “a-e” at 0.002, 0.01, 0.05 and 0.1 mol L<sup>-1</sup> respectively.



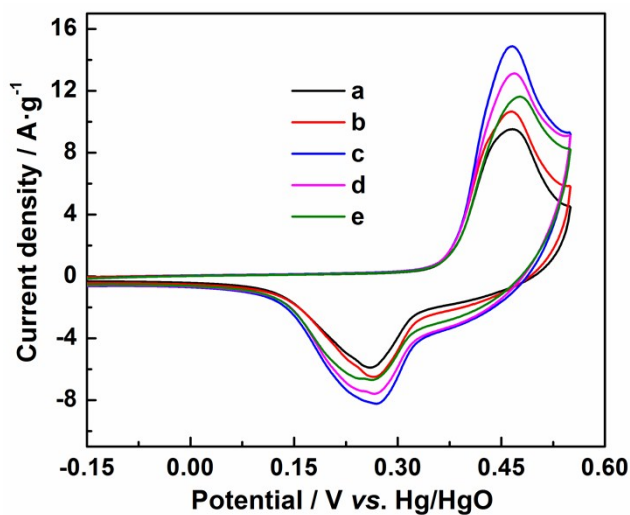
**Fig.S6** Raman spectra of Ni(OH)<sub>2</sub>@Mn<sub>2</sub>O<sub>3</sub> prepared using KMnO<sub>4</sub> concentrations “a-e” at 0.002, 0.01, 0.05 and 0.1 mol L<sup>-1</sup> respectively.



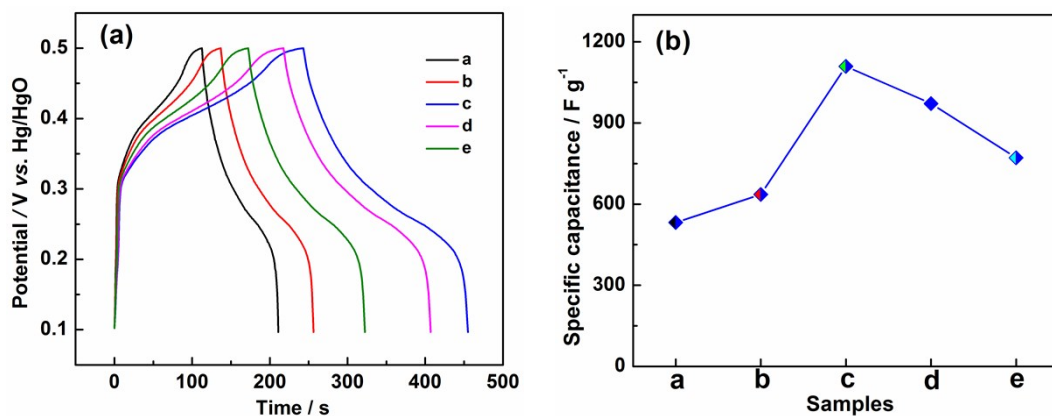
**Fig. S7**  $\text{N}_2$  isotherms of (a)  $\text{Ni}(\text{OH})_2$  and  $\text{Ni}(\text{OH})_2@ \text{Mn}_2\text{O}_3$  prepared using  $\text{KMnO}_4$  concentrations at (b) 0.002, (c) 0.01, (d) 0.02, (e) 0.05 and (f) 0.1  $\text{mol L}^{-1}$  respectively, Inset: the corresponding pore size distribution; and (g) BET surface areas of the six samples.



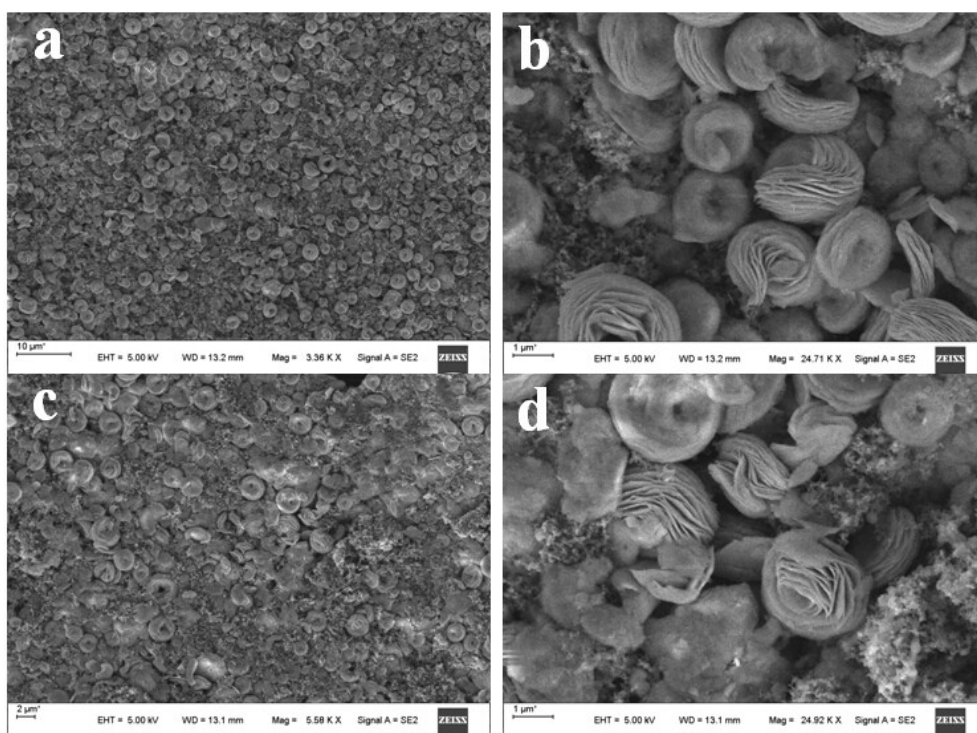
**Fig.S8** XPS of  $\text{Ni(OH)}_2@Mn_2O_3$ . (a) overall XPS survey; (b) Ni 2p; (c) Mn 2p; and (d) O 1s.



**Fig.S9** CV curves of  $\text{Ni(OH)}_2@Mn_2O_3$  prepared using different  $\text{KMnO}_4$  concentrations "a-e" at 0.002, 0.01, 0.02, 0.05 and 0.1 mol L<sup>-1</sup> respectively. Scan rates: 50 mV s<sup>-1</sup>; Electrolyte: 6 mol L<sup>-1</sup> KOH+0.3 mol L<sup>-1</sup> LiOH solution.



**Fig.S10** (a) Galvanostatic charge-discharge plots of Ni(OH)<sub>2</sub>@Mn<sub>2</sub>O<sub>3</sub> and (b) specific capacitance of samples prepared using different KMnO<sub>4</sub> concentrations "a-e" at 0.002, 0.01, 0.02, 0.05 and 0.1 mol L<sup>-1</sup> respectively. Current density: 2 A g<sup>-1</sup>; Electrolyte: 6 mol L<sup>-1</sup> KOH + 0.3 mol L<sup>-1</sup> LiOH solution.



**Fig.S11** SEM images of Ni(OH)<sub>2</sub>@Mn<sub>2</sub>O<sub>3</sub> before (a,b) and after (c,d) 1000 continuous charge-discharge cycles at a current density of 2 A g<sup>-1</sup>.