Experimental section

Synthesis of Mn-oleate complex

The Mn-oleate precursor was prepared by a simple chemical reaction of $MnCl_2$ and sodium oleate, according to the previous report with minor adjustment.[1] 1.6 g of $MnCl_2 \cdot 6H_2O$ and 4.88 g of sodium oleate were first dissolved in a mixture solvent composed of 12 ml H_2O, 16 ml ethanol and 28 ml hexane. The resulting solution was heated to 70 °C and kept at that temperature for four hours. When the reaction was completed, the upper organic layer containing the Mn–oleate complex was washed three times with 30 ml distilled water in a separator funnel. After washing, hexane was evaporated off, resulting in Mn–oleate complex in a waxy solid form.

Synthesis of MnO@graphene/graphene networks

In a typical synthetic procedure, 2 g of the Mn-oleate complex synthesized as described abovewas heated to 700, 850 and 1000 °C with a constant heating rate of 5 °C min^{-1}, and then kept at that temperature for 2 hours. After that, the resulting samples were then cooled to room temperature for further characterizations and electrochemical tests.

Characterizations

The scanning electron microscopy (SEM) images were taken with a JEOLJSM-6700F field emission scanning electron microscope (15 kV). The transmission electron microscopy (HR-TEM) images were taken on a JEOL 2010 high-resolution transmission electron microscope performed at 200 kV. The specimen of HR-TEM measurement was prepared via spreading a droplet of ethanol suspension onto a copper grid, coated with a thin layer of amorphous carbon film, and allowed to dry in air. The X-ray diffraction (XRD) patterns of the products were recorded with Rigaku D/max Diffraction System using a Cu Kα source (λ = 0.15406 nm).Thermal gravimetric analysis (TGA) was performed tobetter understand the decomposition of MnO@G/GNs in oxygen environment with increased temperature. A TG-DTA7300 instrument was used. The composite was heated at 5°C min-1 up to 110°C for 30 min to remove moisture contained in the material and then heated continuously up to 1000°C in argon. The Raman spectra were collected on a Raman spectrometer (Labram-010) using 632nm laser. The thermogravimetric analysis was carried out by a STA449C instrument with a heating rate of 5 °C in Ar. X-ray photoelectron spectroscopic (XPS) measurements were performed on an ESCALAB 250Xi using a monochromic Al X-ray source (200 W, 20 eV).

Electrochemical tests

The working electrodes were prepared by mixing 80 wt% as-prepared active materials with 10 wt% carbon black and 10 wt% carboxymethylcellulose (CMC) binder in distilled water to form homogeneous slurry, which was uniformly pasted onto copper foil. The prepared working electrodes were dried in a vacuum oven at 80 °C over 12 h and were then ready for assembly in test cells after pressing. Electrochemical cells (CR2032 coin type) using the active materials (about 1.5 mg per sheet) as working electrode, Li foil as the counter electrode and reference electrode, a microporous polypropylene film as the separator, and 1 M $LiPF_6$ in a 1:1 (v/v) mixture of ethylene carbonate (EC) and dimethyl carbonate (DMC) as the electrolyte were assembled in an Ar-filled glove box (H_2O, $O_2 < 0.1$ ppm, Mbraun, Unilab, USA). The cells were galvanostatically charged and discharged over a voltage range of 0.02-3 V versus Li/Li^+ at different constant current densities based on the weight of the samples on a Land CT2001A cycler. Cyclic voltammetry (CV) was performed on an Ametek PARSTAT®2273 electrochemistry workstation.

Reference

1   T. Y. Wang, Z. Peng, Y. H. Wang, J. Tang and G. F. Zheng, *Sci. Rep.*, **2013**, *3*, 2693.
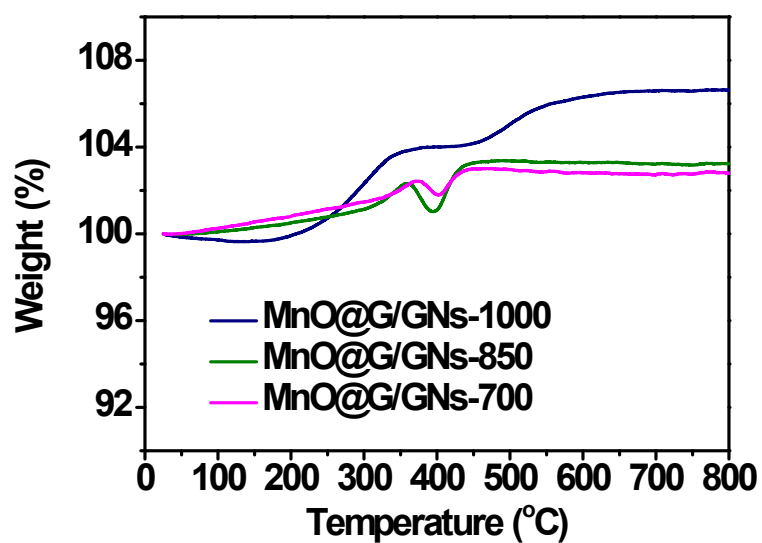
Figure S1 TGA curves of MnO@G/GNs.

The content of carbon in the composites can be calculated, based on the integrative effect of the weight loss (arising from the combustion of carbon to CO_2) and the weight gain (arising from the oxidation of MnO to Mn_2O_3).[1,2]

References

1   W. J. Zhu, H. Huang, W. K. Zhang, X. Y. Tao, Y. P. Gan, Y. Xia, H. Yang and X. Z. Guo, *Electrochim. Acta*, **2015**, *152*, 286-293.

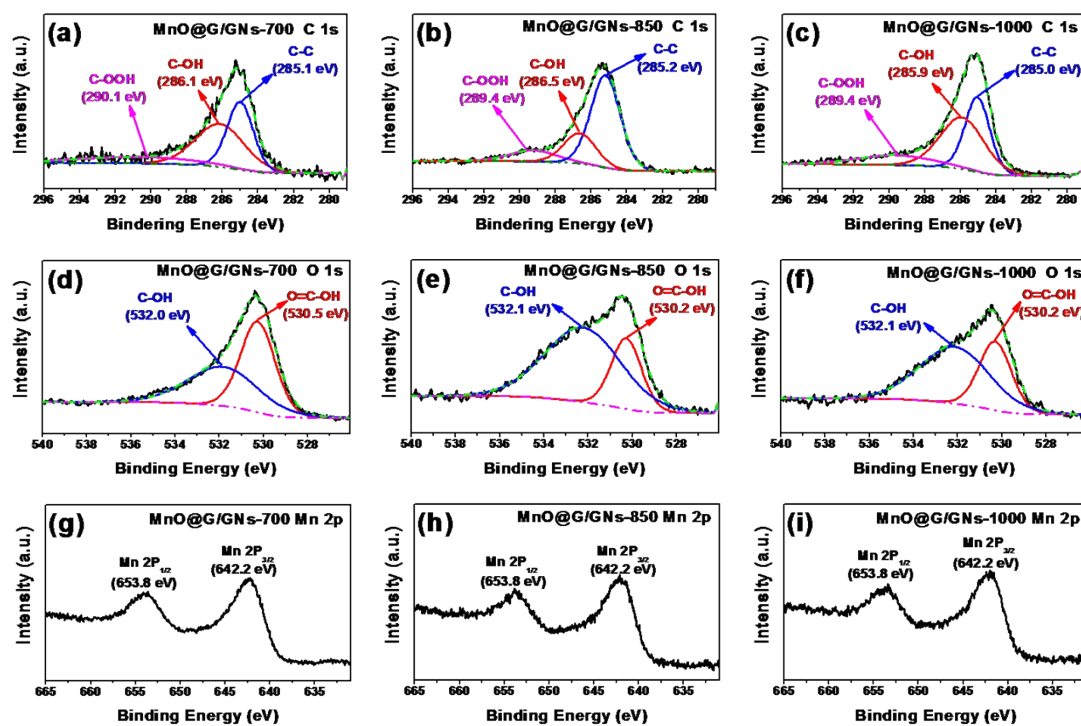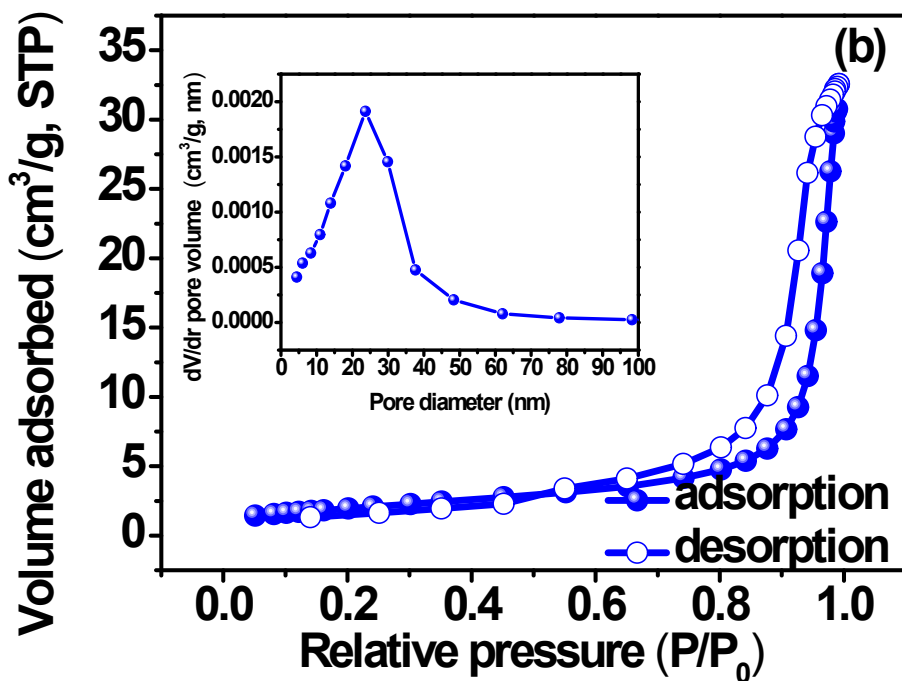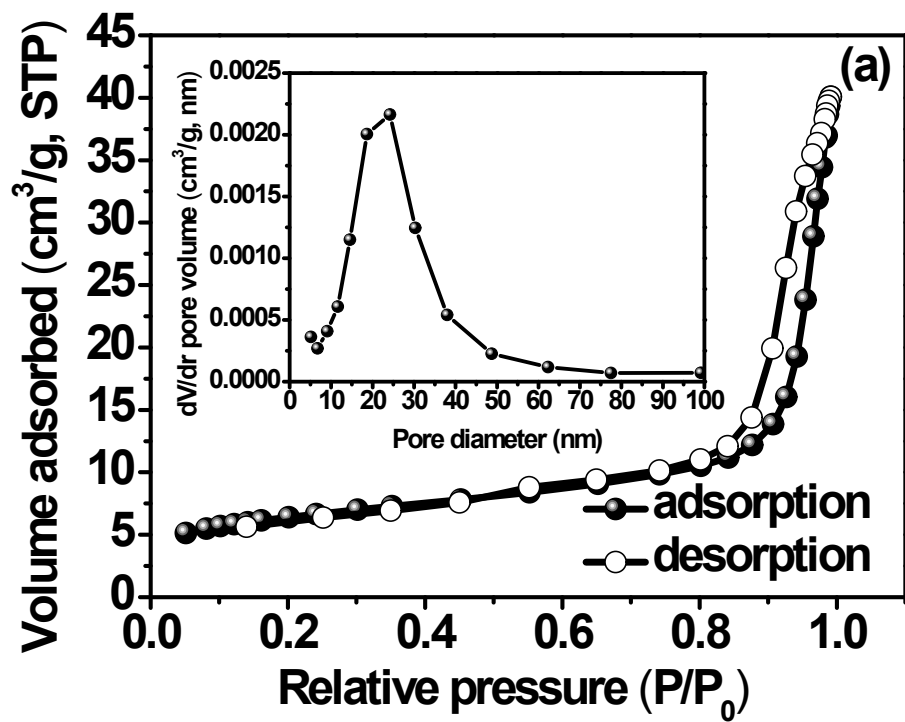2   Y. M. Sun, X. L. Hu, W. Luo, F. F. Xia and Y. H. Huang, *Adv. Funct. Mater.*, **2013**, *23*, 2436–2444.

Figure S2 XPS spectra of MnO@G/GNs: (a-c) C 1s; (d-f) O 1s; (g-i) Mn 2p.

Figure S3 (a and b)Nitrogen absorption-desorption isotherms and pore size distribution (inset) of MnO@G/GNs-700 and MnO@G/GNs-1000.
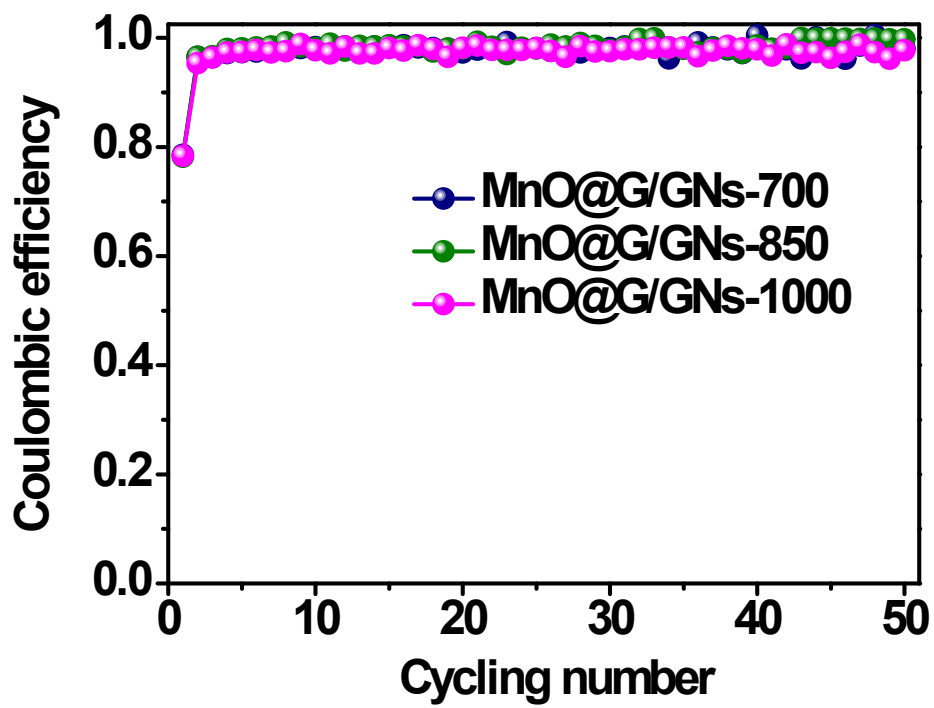
Figure S4 Coulombic efficiency of MnO@G/GNs during cycling at 50 mAh/g.

Table S1 Typical MnO/carbon composites and their electrochemical performance

Structures	Carbon content	Performances	References
core-shell	19.6 wt%	952 mAh/g after 200 cycles at 0.2 A/g	14
Chinese lantern-like MnO@N–C	23.8 wt%	640 mAh/g after 400 cycles at 0.2 A/g	15
MnO@C nanorods	43.1 wt%	481 mAh/g after 50 cycles at 0.2 A/g	16
core–shell nanoplates	13.7 wt%	770 mAh/g after 30 cycles at 0.2 A/g	19
MnO/graphene	17.4 wt%	2014.1 mAh/g after 150 cycles at 0.2 A/g	22
MnO/graphene	14.57%	730 mAh/g after 50 cycles at 0.1 A/g	24
3D porous MnO/C microspheres	5.71 wt%	864 mAh/g^{-1} after 100 cycles at 0.1 A/g	25