

## ***Supplementary Information***

### One Dimensional Graphene Nanoscroll-Wrapped MnO Nanoparticles for High-Performance Lithium Ion Hybrid Capacitors

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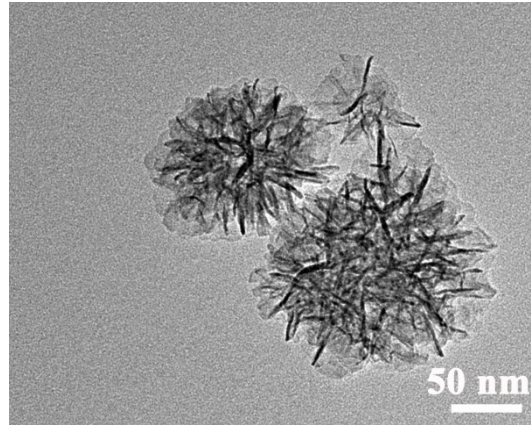
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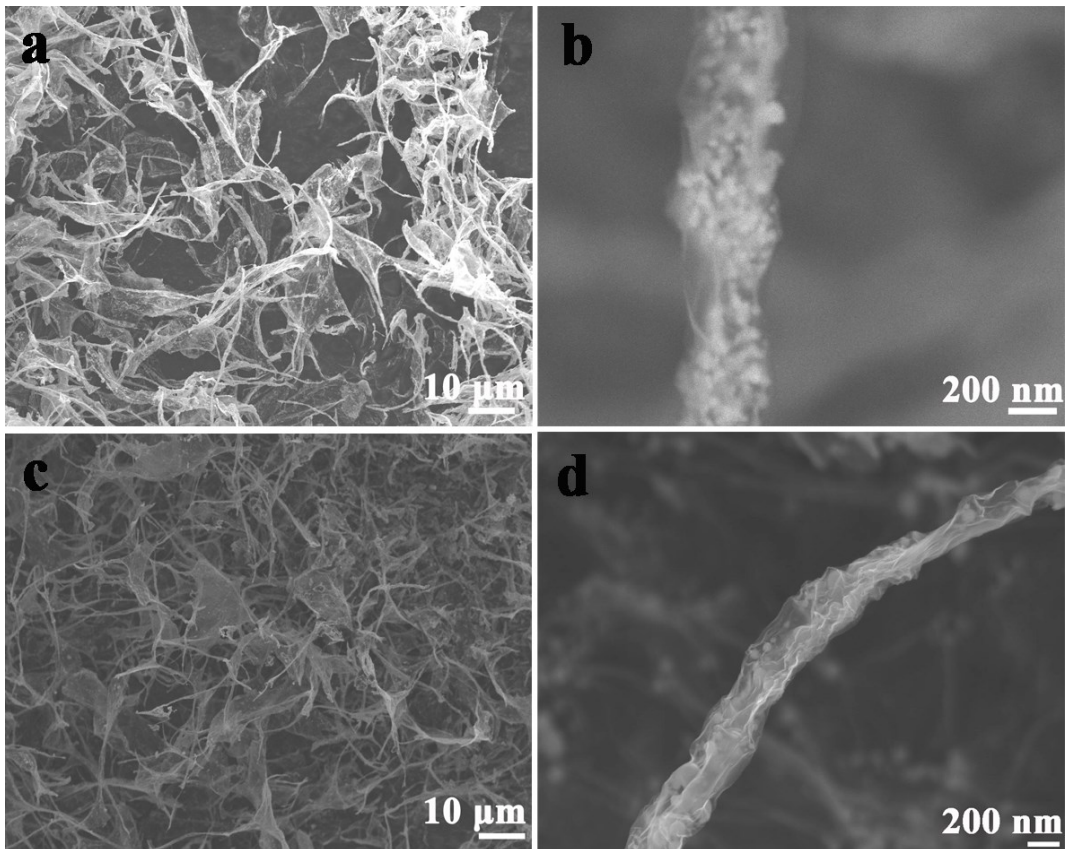
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Keywords: graphene nanoscroll, MnO, anode, 3D framework activated carbon, lithium-ion hybrid capacitors.



**Figure S1.** High magnification TEM image of spherical  $K_xMnO_2$  precursors.



**Figure S2.** High and low magnification SEM images of GNS@MnO with different  
Calcination temperature: (a,b) GNS@MnO<sub>x</sub>-400, (c,d) GNS@MnO-800.

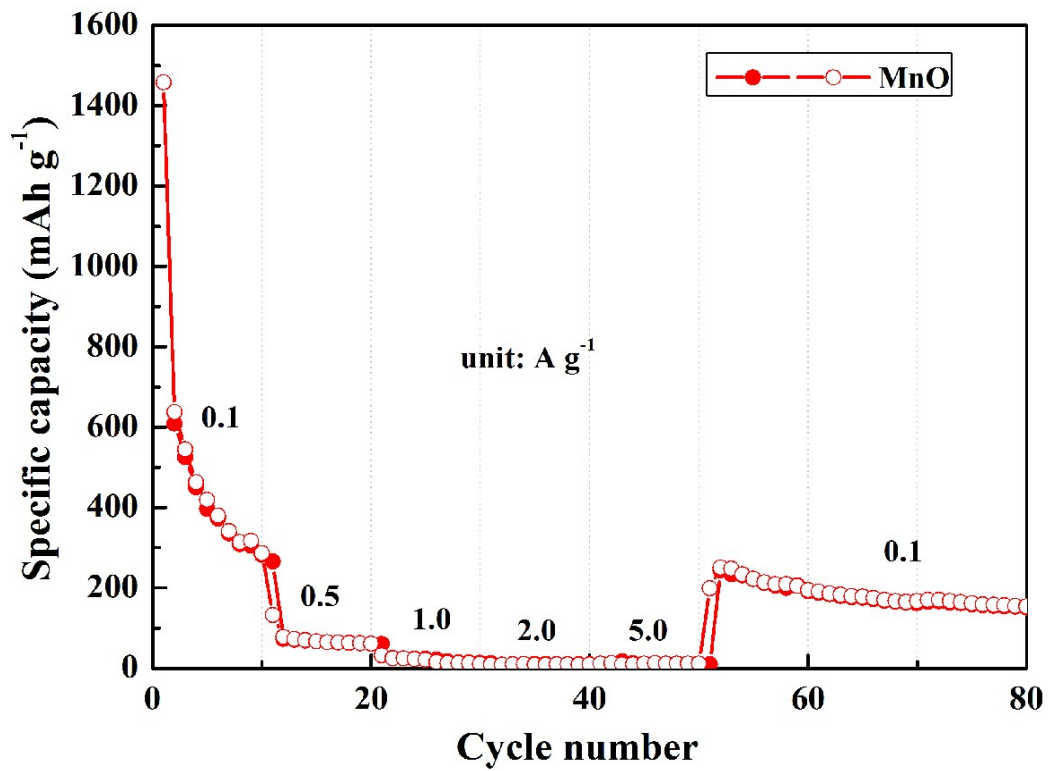


Figure S3. Rate performance of pure MnO electrode.

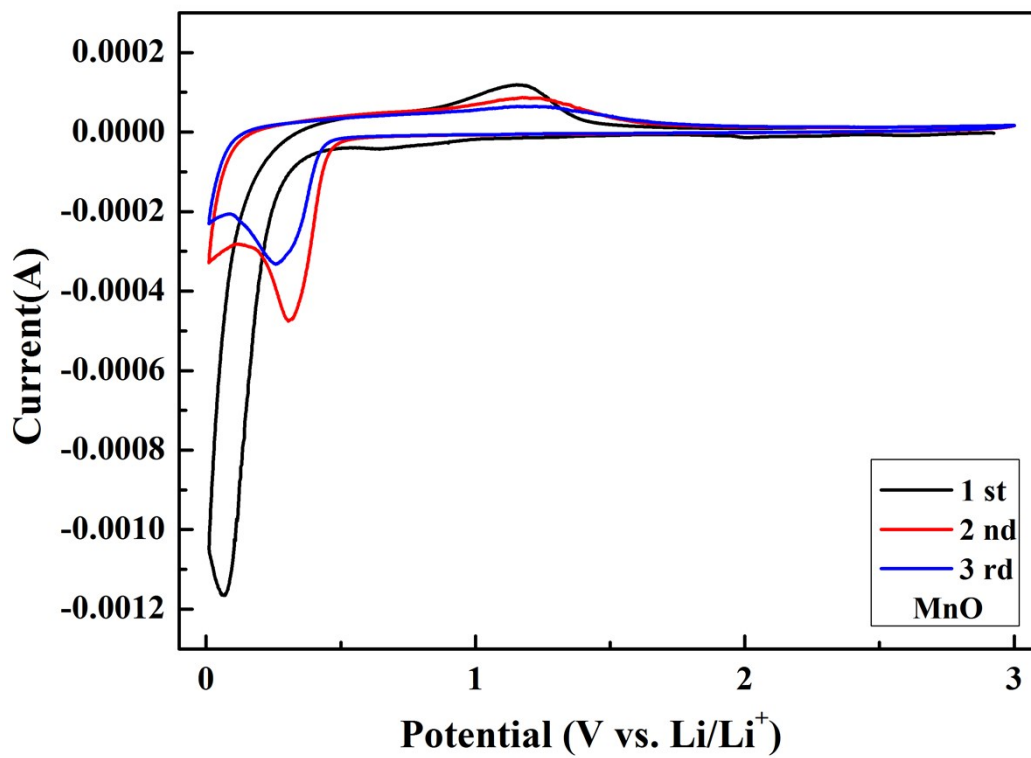
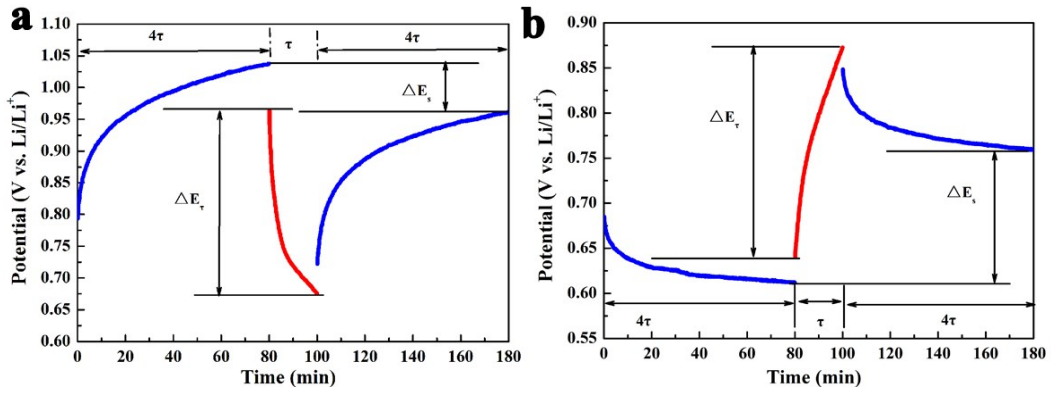
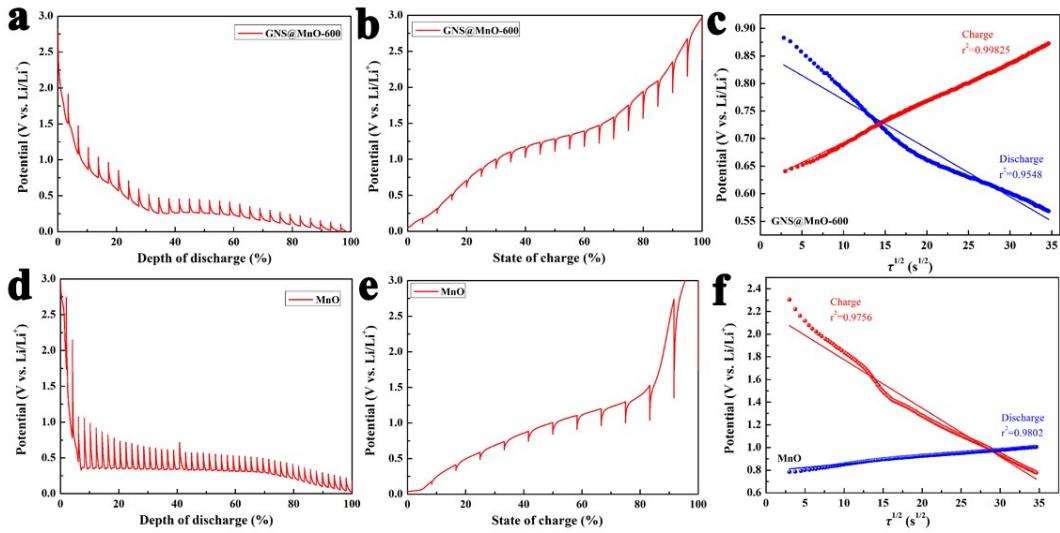


Figure S4. The first three CV curves of MnO at a scan rate of 0.1 mV s<sup>-1</sup>.



**Figure S5.** Scheme for voltage response with time during a single constant current pulse: (a) Discharge. (b) Charge.



**Figure S6.** GITT characterization: GITT potential profiles of (a) discharge and (b) charge for GNS@MnO-600 anode. (c) Variation of cell potential during single charge and discharge titration plotted against  $\tau^{1/2}$  for GNS@MnO-600 anode. GITT potential profiles of (d) discharge and (e) charge for MnO anode. (f) Variation of cell potential during single charge and discharge titration plotted against  $\tau^{1/2}$  for pure MnO anode.

GITT is an effective strategy to estimate the apparent  $\text{Li}^+$  ion diffusion coefficient at different quasi-equilibrium potentials. During the test, a relatively small titration current density (i.e.,  $0.1 \text{ A g}^{-1}$ ) is applied for a relatively short period (i.e.,  $\tau = 20 \text{ min}$

= 1200 s) to induce a potential shift ( $\Delta E\tau$ ), followed by much longer relaxation period (i.e.,  $4\tau = 80 \text{ min} = 4800 \text{ s}$ ) to reach a quasi-equilibrium potential for the calculation of  $\Delta E_s$  (Figure S5). The above titration-relaxation cycle is performed continuously at the whole potential window (i.e., 0.01-3.0 V vs. Li/Li<sup>+</sup>) to give a completed potential profile (Figure S6).

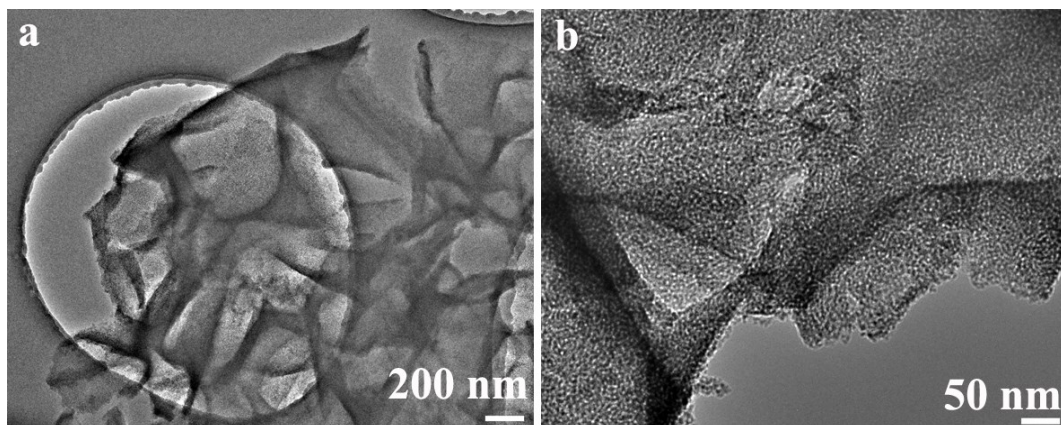
The apparent Li<sup>+</sup> diffusion coefficient ( $D$ , cm<sup>2</sup> s<sup>-1</sup>) is calculated based on Fick's second law of diffusion (equation S1):<sup>1,2</sup>

$$D = \frac{4}{\pi} \left( \frac{mV_m}{MA} \right)^2 \left( \frac{\Delta E_s/\tau}{dE_\tau/d\sqrt{\tau}} \right)^2 \quad (\text{S1})$$

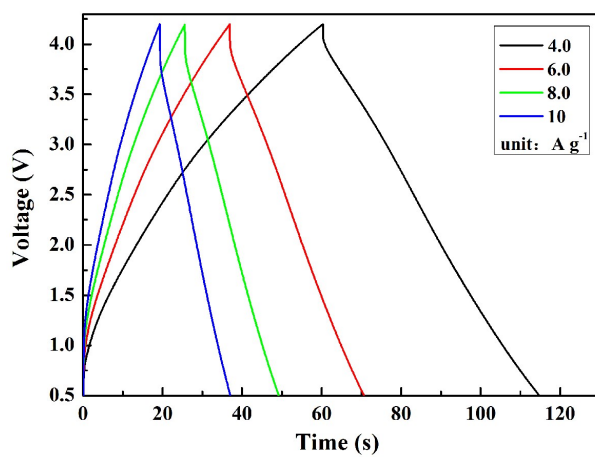
where  $m$ (g) is mass loading,  $V_m$  (cm<sup>3</sup> mol<sup>-1</sup>) is molar volume of the electrode,  $M$  (g mol<sup>-1</sup>) is molar weight of the electrode,  $A$  (cm<sup>2</sup>) is electroactive area of the electrode,  $\Delta E_s$  (V) is the change of quasi-equilibrium potential after two sequential relaxation period,  $\tau$  (s) is charge or discharge time during each titration,  $dE_\tau/d\sqrt{\tau}$  (V s<sup>-1/2</sup>) is potential shift rate.

The above equation S1 can be simplified as equation S2 by applying the small current density for a sufficiently short time in each titration.

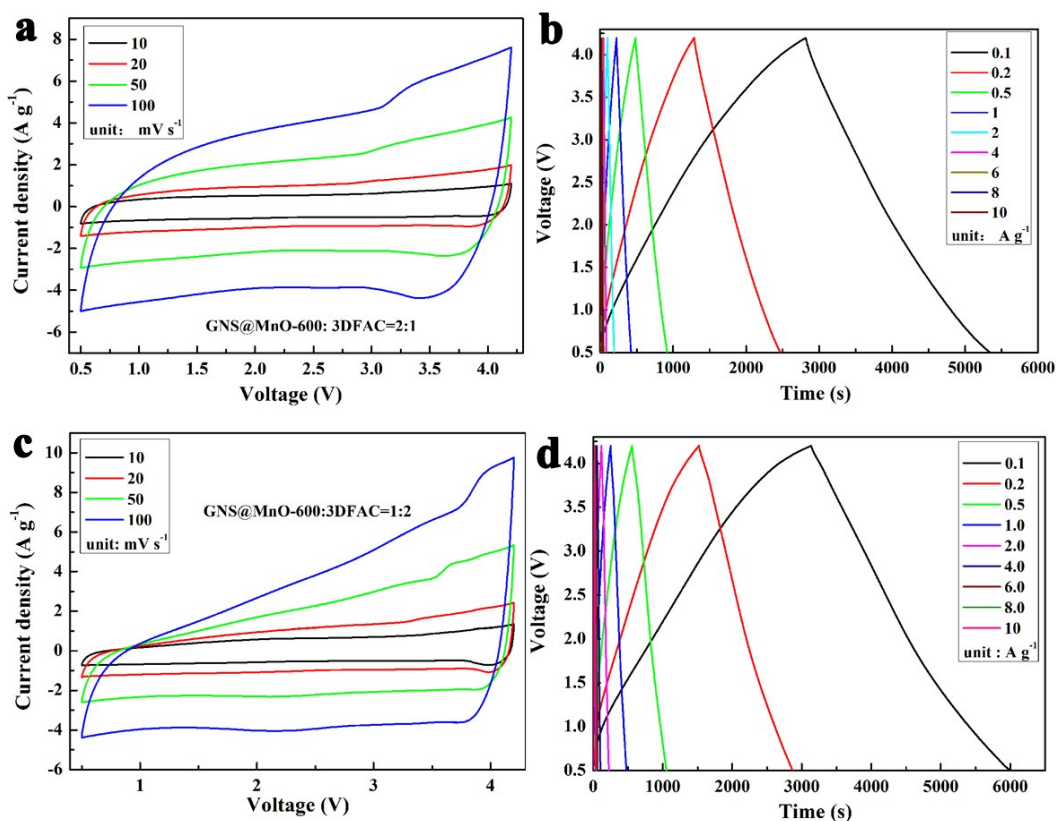
$$D = \frac{4}{\pi\tau} \left( \frac{mV_m}{MA} \right)^2 \left( \frac{\Delta E_s}{\Delta E_\tau} \right)^2 \quad (\text{S2})$$



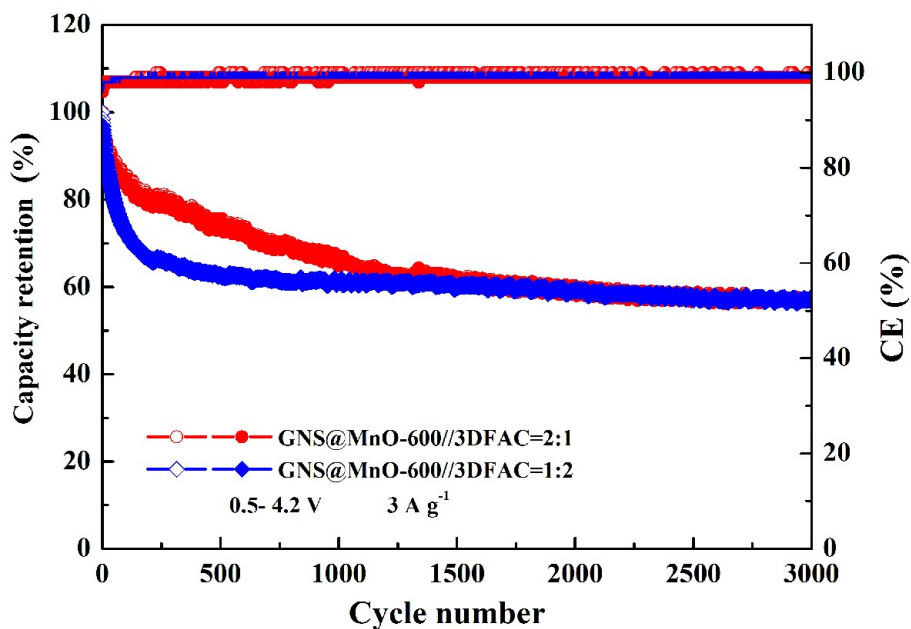
**Figure S7.** TEM images of 3DFAC



**Figure S8.** GCD profiles of GNS@MnO-600//3DFAC LIHC (1:1) at high current density.



**Figure S9.** CVs and GCD profiles of GNS@MnO-600//3DFAC LIHCs with different mass ratios: (a, b) 2:1; (c, d) 1:2.



**Figure S10.** Cycling stability of the as-assembled GNS@MnO-600//3DFAC devices with mass ratios of 2:1 and 1:2.

Table S1. Comparison of the electrochemical performances of the as-prepared 1D GNS@MnO-600 anode with other MnO-C anode materials for LIBs reported previously.

Materials	Rate performance	Ref.
MnO@NC/ graphite	0.1 A g <sup>-1</sup> / 835 mA h g <sup>-1</sup> 5.0 A g <sup>-1</sup> / 387 mA h g <sup>-1</sup>	3
NC/MnO/RGO	0.1 A g <sup>-1</sup> / 925.9 mA h g <sup>-1</sup> 5.0 A g <sup>-1</sup> / 355.5 mA h g <sup>-1</sup>	4
MnO@HCF-2	0.1 A g <sup>-1</sup> / 586.8 mA h g <sup>-1</sup> 4.0 A g <sup>-1</sup> / 327.8 mA h g <sup>-1</sup>	5
MnO@N-CS	0.2 A g <sup>-1</sup> / 917mA h g <sup>-1</sup> 5.0 A g <sup>-1</sup> / 328 mA h g <sup>-1</sup>	6
MnO/Mn3O4/N-graphene	0.5 A g <sup>-1</sup> / 681 mA h g <sup>-1</sup> 2.0 A g <sup>-1</sup> / 365 mA h g <sup>-1</sup>	7
MnO/N-PCNTs	0.1 A g <sup>-1</sup> / 652 mA h g <sup>-1</sup> 1.0 A g <sup>-1</sup> / 220 mA h g <sup>-1</sup>	8
MnO/Metal/Carbon	0.1 C / 600 mA h g <sup>-1</sup> 5.0 C / 200 mA h g <sup>-1</sup>	9
Sn-MnO@CYINs	0.2 A g <sup>-1</sup> / 662 mA h g <sup>-1</sup> 2.0 A g <sup>-1</sup> / 402 mA h g <sup>-1</sup>	10
RGO-MnO@NC	0.5 A g <sup>-1</sup> / 599 mA h g <sup>-1</sup> 5.0 A g <sup>-1</sup> / 331 mA h g <sup>-1</sup>	11
C/MnO/SiOC	0.1 A g <sup>-1</sup> / 684 mA h g <sup>-1</sup> 2.0 A g <sup>-1</sup> / 408 mA h g <sup>-1</sup>	12
MnO-PS	0.1 A g <sup>-1</sup> / 960 mA h g <sup>-1</sup> 2.0 A g <sup>-1</sup> / 300 mA h g <sup>-1</sup>	13
<b>1D GNS@MnO-600</b>	<b>0.1 A g<sup>-1</sup> / 766 mA h g<sup>-1</sup></b> <b>5.0 A g<sup>-1</sup> / 437 mA h g<sup>-1</sup></b>	<b>This work</b>

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