Support Information

Control over the morphology and phase of MnO_x formed in the modified Hummers method and impact on the electrocapacitive properties of MnO_x-graphite oxide composite electrodes

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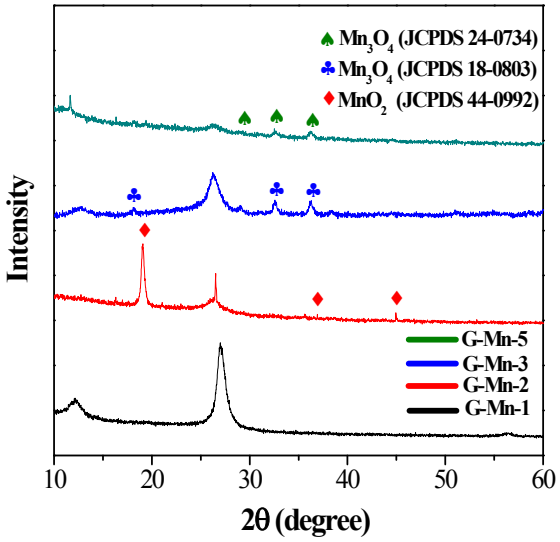

Figure S1. XRD patterns of as prepared MnO_x-graphite oxide composites.

All samples exhibit a peak at around 26.6 degrees two theta, which is attributed to graphite oxide. No peaks due to manganese oxide can be seen from G-Mn-1. This may be due to the low content of manganese oxide and/or low crystallinity. The peaks that are observed from G-Mn-2 can be indexed to λ-MnO_2 (JCPDS 44-0992). The peaks seen from G-Mn-3 are due to diffraction of γ-Mn_3O_4 according to JCPDS 18-0803.[1] The peaks seen from G-Mn-5 are due to diffraction of tetragonal hausmannite-Mn_3O_4 (JCPDS 24-0734).[2] These results confirmed the presence of different MnO_x phases on the graphite oxide surface and matching well with the XPS results in Figure 3.

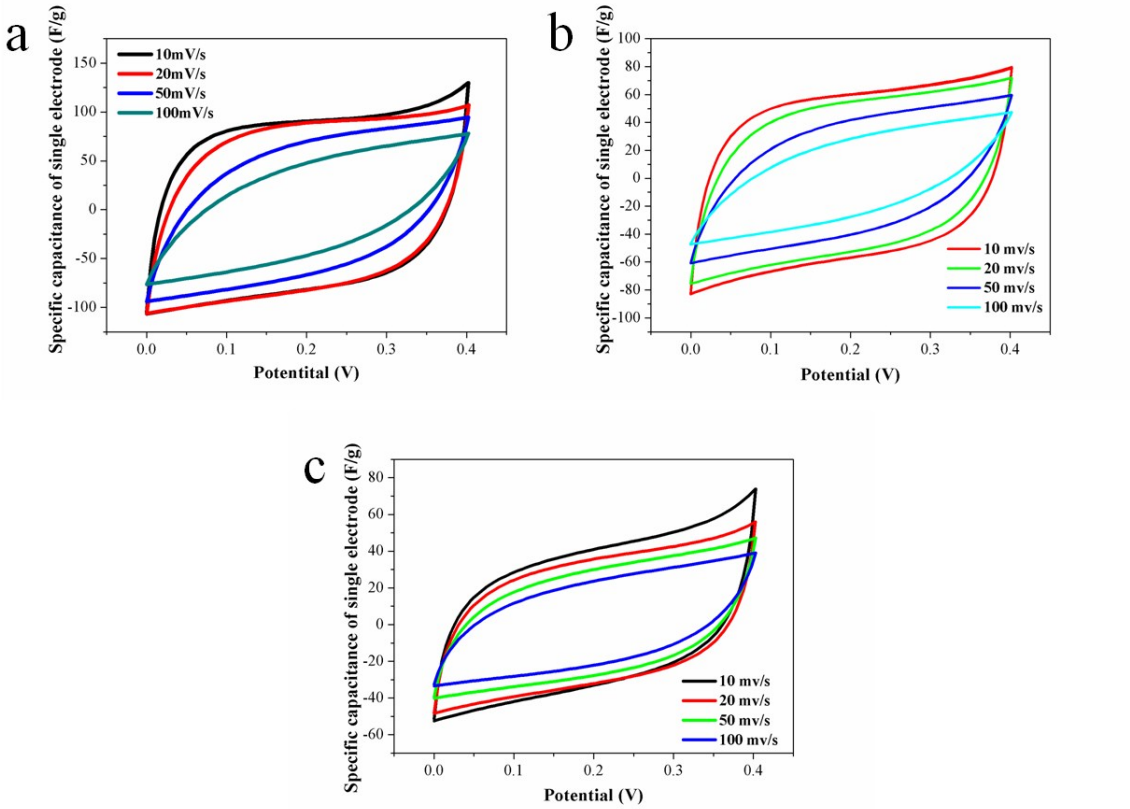

Figure S2. CV curves of G-Mn-1 (a), G-Mn-3 (b), G-Mn-5 (c).

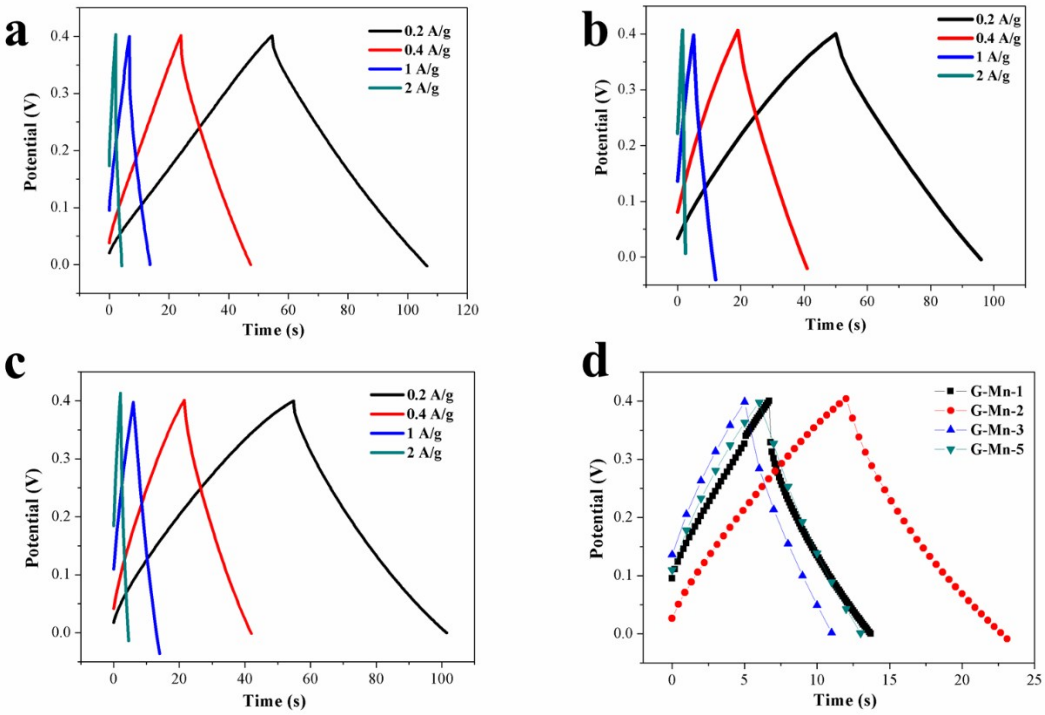

Figure S3. GCD curves of G-Mn-1 (a), G-Mn-3 (b), and G-Mn-5 (c) at different current rates and GCD curves of samples at a current rate of 1 A/g (d).

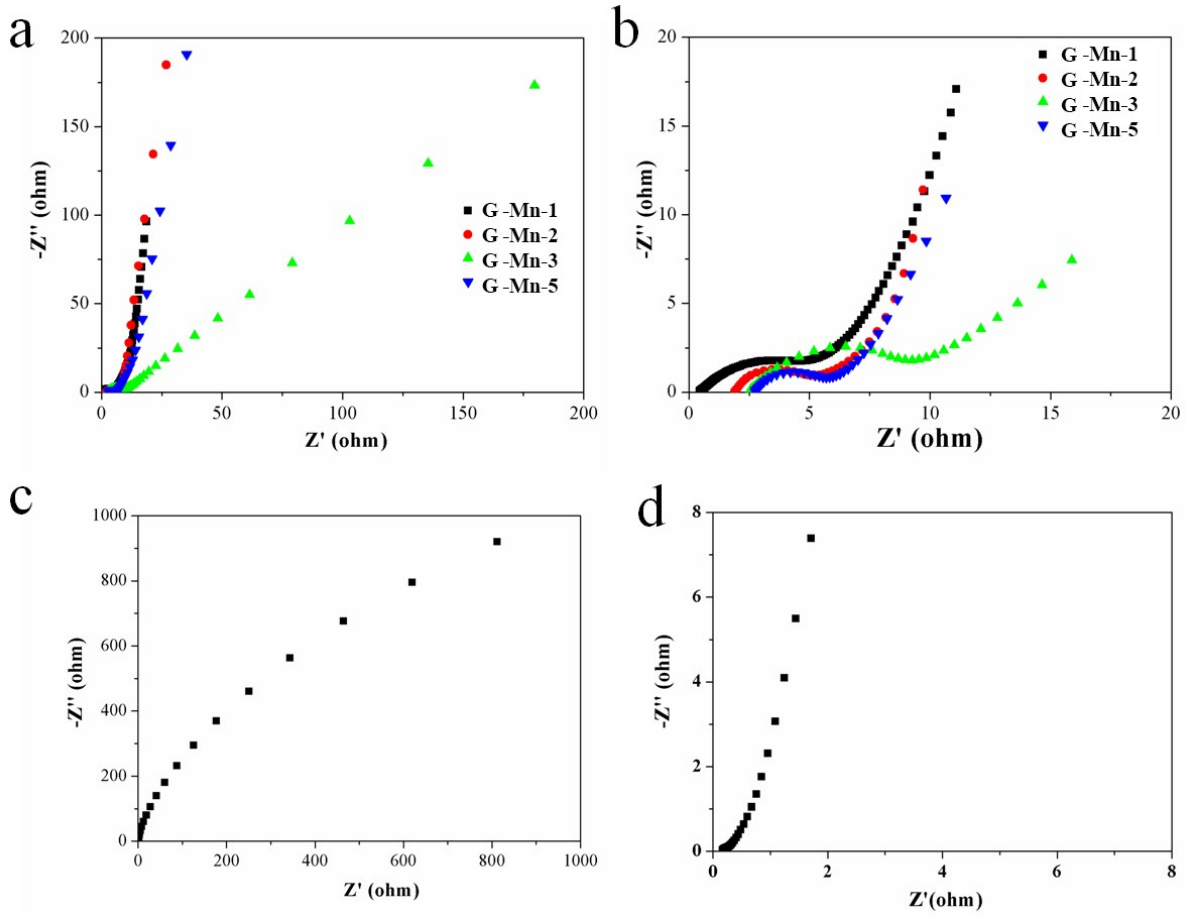

Figure S4. EIS comparison of (a) MnO_x-graphite oxide composites with (b) corresponding enlarged EIS data and (c) pristine graphite oxide with (d) corresponding enlarged EIS data.

Figure S4 shows the Nyquist plots of four MnO_x-graphite oxide composites (a, b) and pristine graphite oxide (c, d). They all share semicircle in the high frequency region followed by inclined lines in the low frequency region. The semicircle implies the charge transfer resistance and the intersection at the real axis indicates the equivalent series resistance (ESR). The ESR values of G-Mn-1, G-Mn-2, G-Mn-3, G-Mn-5 obtained from the Nyquist plots are 0.5 Ω, 1.9 Ω, 2.5 Ω and 2.8 Ω, respectively. The ESR value of the pristine graphite oxide is 0.16 Ω. G-Mn-2 shows a more vertical line in Warburg resistance corresponding to an electrode closer to an ideal capacitor.[3]

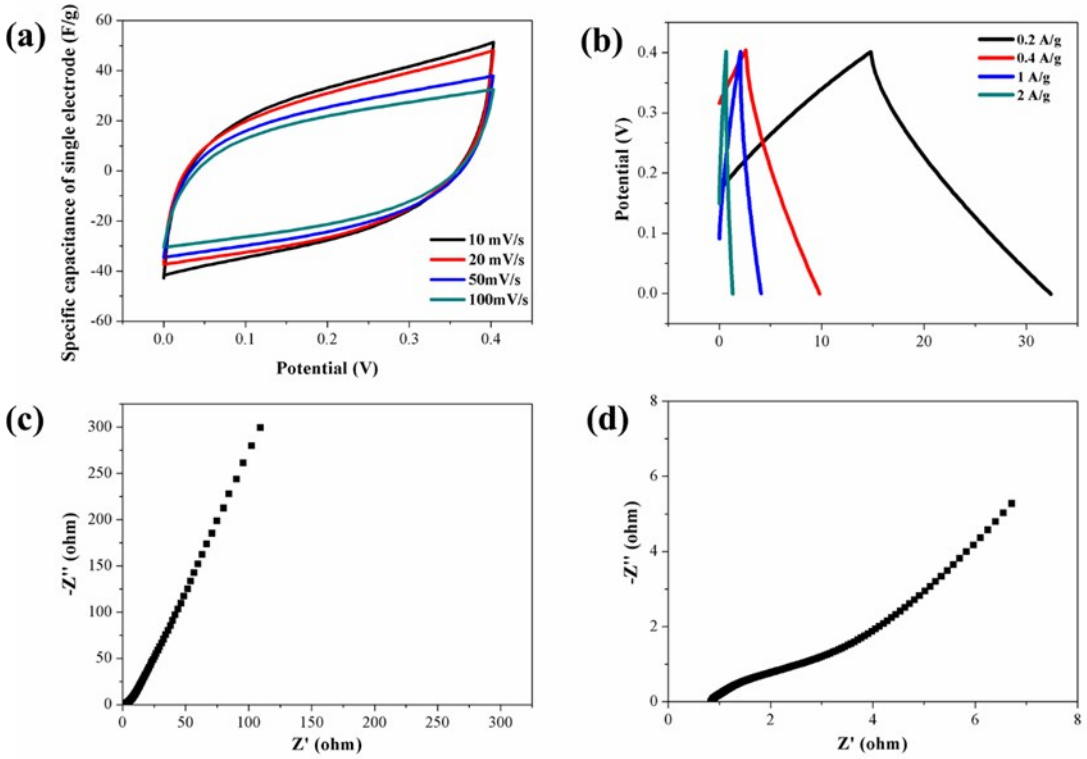

Figure S5. Electrochemical performance of G-Mn-2 in 1 M Na_2SO_4 solution. (a) CV curves, (b) GCD curves, (c) EIS profile, and (d) high-frequency region EIS profile.

[1] A. A. Badawy and S. M. Ibrahim, *International Journal of Industrial Chemistry*, 2016, DOI: 10.1007/s40090-015-0068-2, 1-10.

[2] W. Xiao, J. S. Chen and X. W. Lou, *Cryst Eng Comm*, 2011, 13, 5685-5687.

[3] Y. R. Nian and H. Teng, *Journal of The Electrochemical Society*, 2002, **149**, A1008-A1014.