

Supporting Information

3D MnO₂/Graphene composites with large areal capacitance for high-performance asymmetric supercapacitors

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Data analysis

The power density and energy density were calculated by using the following equations:

$$P = V^2 / [4RS] \quad (1)$$

$$E = 0.5CV^2/S \quad (2)$$

where V is the applied voltage, R is the internal resistance calculated by equation (3), S is the total area of active electrode materials, and C is the measured total areal capacitance of electrode calculated by equation (4):

$$R = \Delta V_{iR} / 2I \quad (3)$$

$$C = I\Delta t / [SV] \quad (4)$$

where I is the applied current, ΔV_{iR} is the voltage drop between the first two points from its top cut-off of discharge curve, Δt is the discharge time after the initial IR drop.

As for the calculation of ASC, the area (S) should be replaced by volume (v).

As for a ASC, the charge balance will follow the relationship $q^+ = q^-$:

$$q = C \times E \times m \quad (5)$$

and in order to get $q^+ = q^-$, the mass balancing will follow the Equation (6):

$$m^+/m^- = C^-E^- / C^+E^+ \quad (6)$$

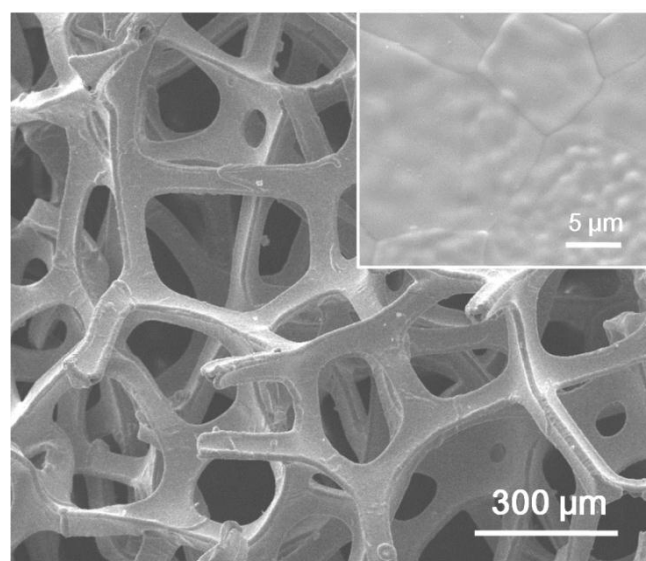


Fig. S1 SEM images of pristine Ni foams.

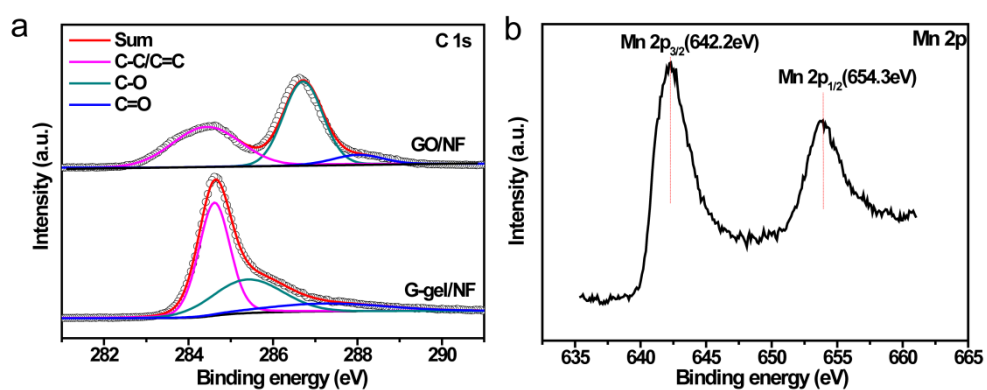


Fig. S2 (a) Normalized C 1s core level XPS spectra of GO/NF and G-gel/NF.

(b) Normalized Mn 2p core level XPS spectrum of MnO₂/G-gel/NF.

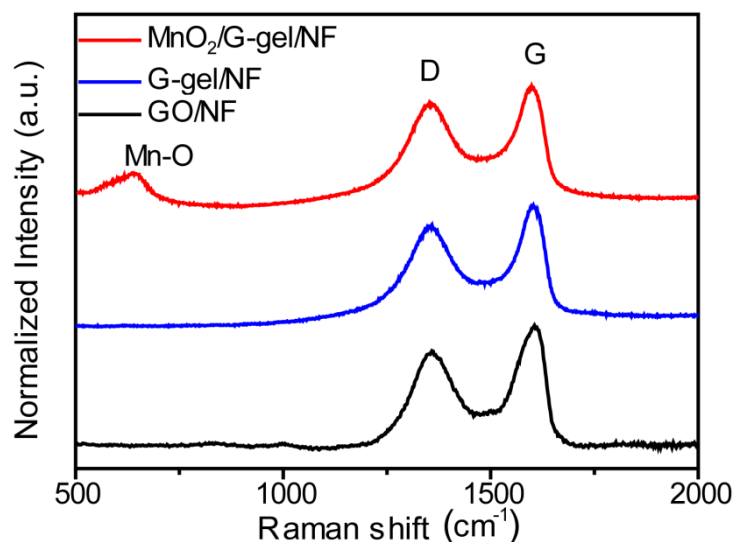


Fig. S3 Raman spectra of GO/NF, G-gel/NF and MnO₂/G-gel/NF.

Fig. S3 shows the typical Raman spectra of GO/NF and G-gel/NF and MnO₂/G-gel/NF. The three electrodes all present two prominent bands around 1353 cm⁻¹ and 1595 cm⁻¹, which can be assigned to the D and G bands of carbon, respectively.^{1, 2} The D band is an indication of the structural defects or partially disordered structures of graphitic domains while the G band is related to the graphitic carbon.³ The I_D/I_G value of GO/NF was calculated to be 0.76 while that of G-gel/NF or MnO₂/G-gel/NF was higher, 0.87. This result indicates that the oxidized areas of GO sheets were restored during hydrothermal process, forming small conjugated domains, again confirming the transformation of GO to graphene.^{3, 4}

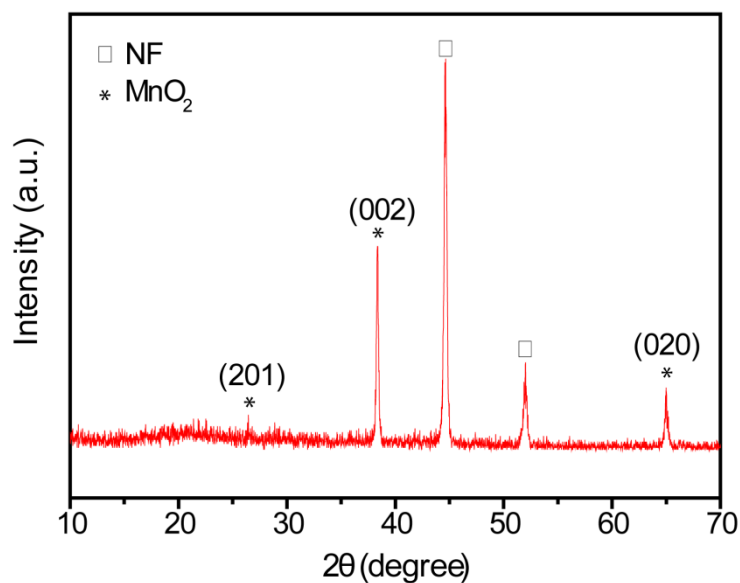


Fig. S4 XRD pattern of MnO₂/G-gel/NF.

Besides the Ni peaks coming from the Ni Foam, three peaks at $\theta = 26.5^\circ$, 38.3° and 65.1° which can be indexed as (201), (002), and (020) of the ramsdelite MnO₂ (JCPDF #42-1316), which is in agreement with the result of the SAED analysis. The weak diffraction point of the (202) and (301) in SAED make it difficult to be indexed in the XRD pattern. It should be noted that no diffraction peak from graphene is observed. This may be due to that the signal of the graphene is much lower than the strong peaks from Ni foam.⁵⁻⁷

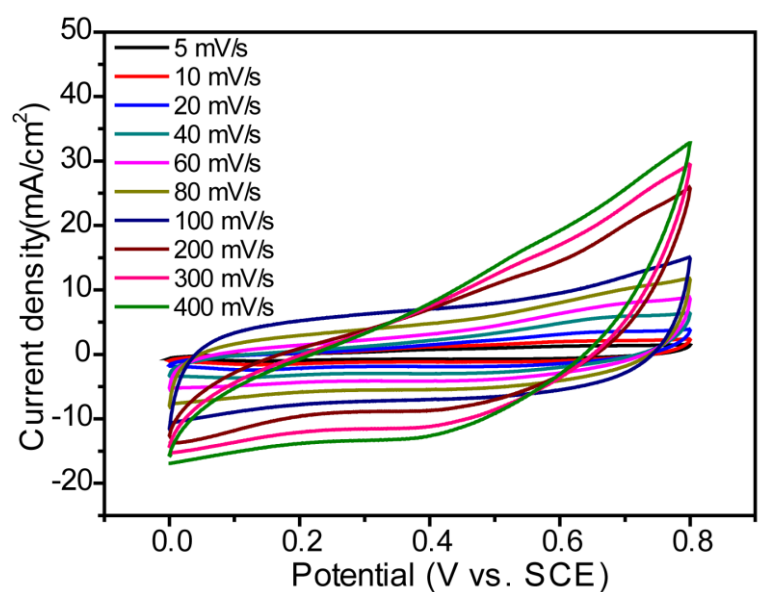


Fig. S5 CV curves of MnO₂/NF collected at different scan rates.

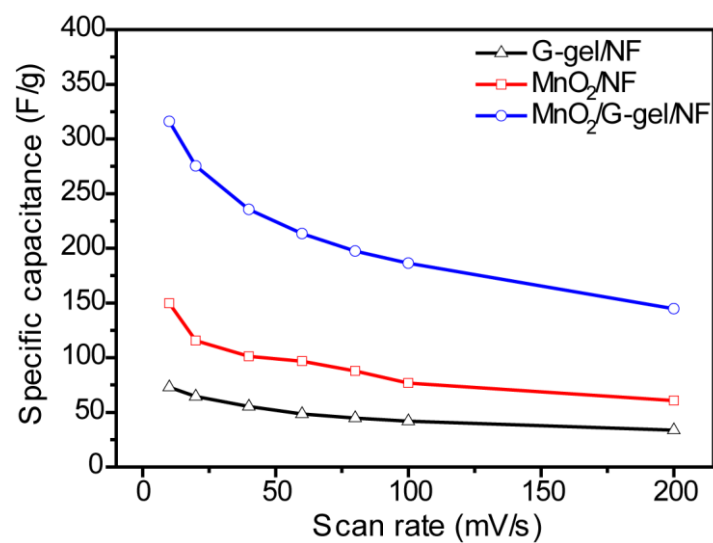


Fig. S6 Specific capacitances of G-gel/NF, MnO₂/NF and MnO₂/G-gel/NF electrodes calculated from CV curves as a function of scan rates.

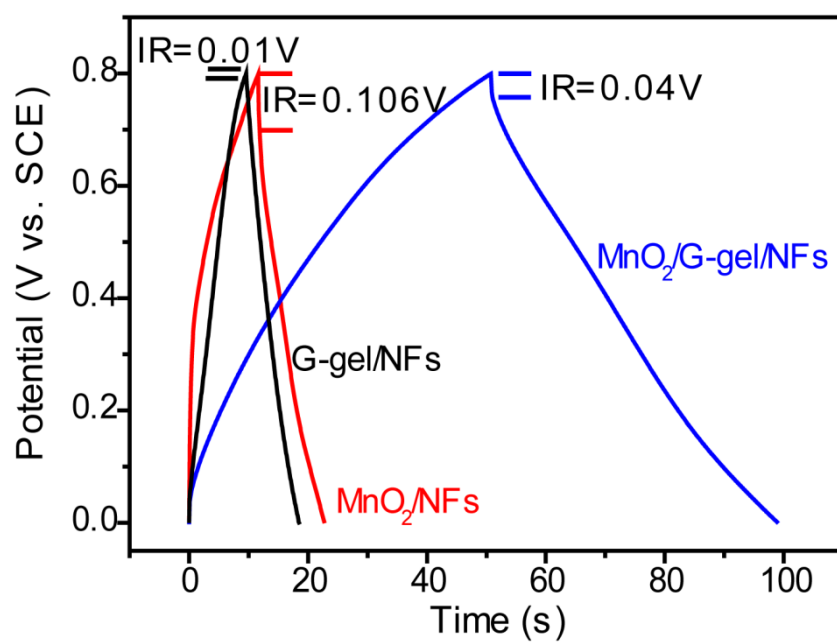


Fig. S7 Galvanostatic charge/discharge curves of MnO₂/NF and MnO₂/G-gel/NF electrodes collected at a current density of 5 A/g. The deposition time of MnO₂ on NF and G-gel/NF was 60 s.

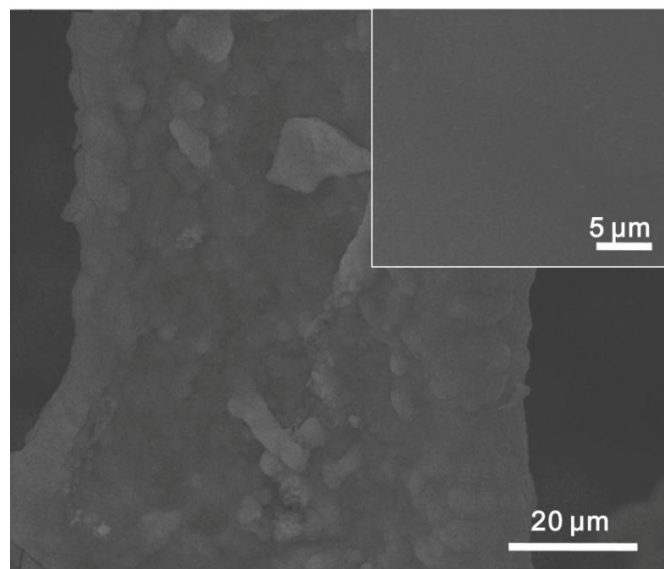


Fig. S8 SEM images MnO₂/G-gel/NF with MnO₂ mass loading of 13.61 mg/cm².

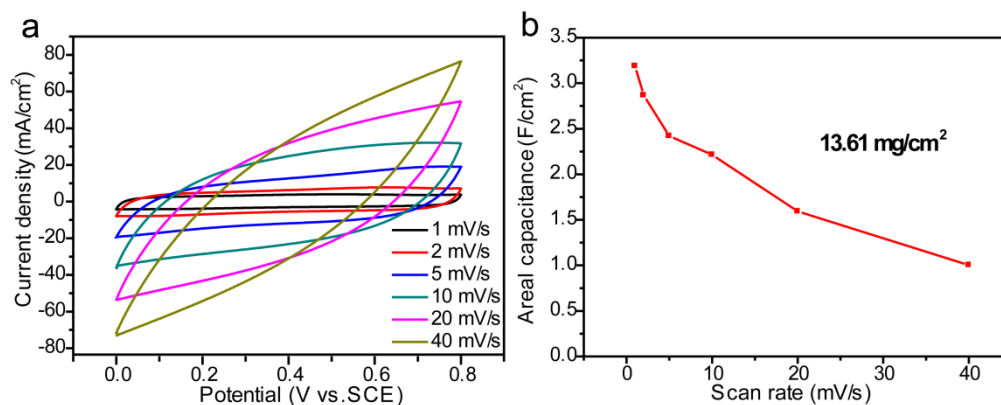


Fig. S9 (a) CV curves of MnO₂ with areal mass loading of 13.61 mg/cm² on G-gel/NF at varies scan rates from 1~40 mV/s. (b) Areal capacitance of MnO₂/G-gel/NF as a function of scan rates.

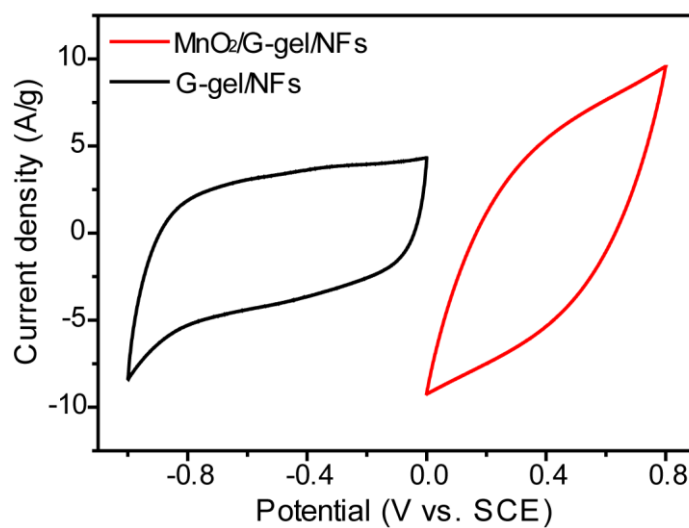


Fig. S10 Comparative CV curves of MnO₂/G-gel/NF and G-gel/NFs performed in a three-electrode cell in 0.5 M Na₂SO₄ aqueous solution at a scan rate of 40 mV/s.

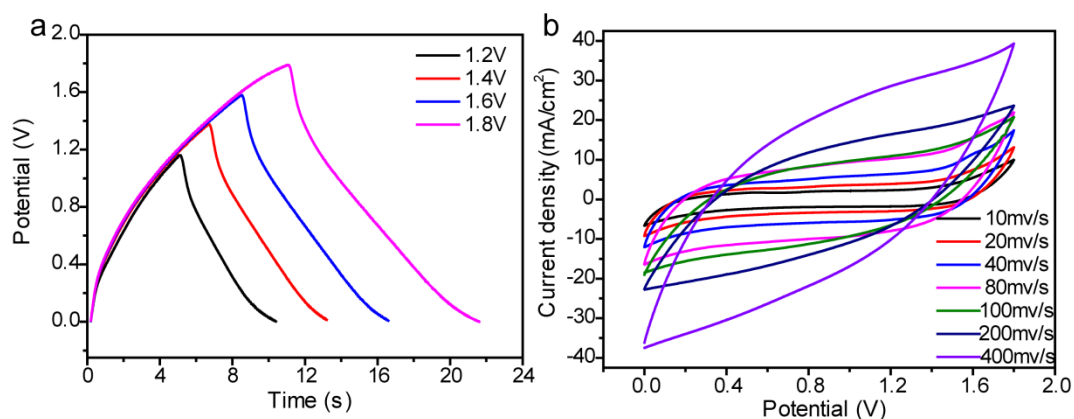


Fig. S11 (a) Galvanostatic charge/discharge curves of as assembled ASC collected at 12 mA/cm² in different potential windows. (b) CV curves of as assembled ASC collected at varied scan rates in a voltage window of 1.8 V.

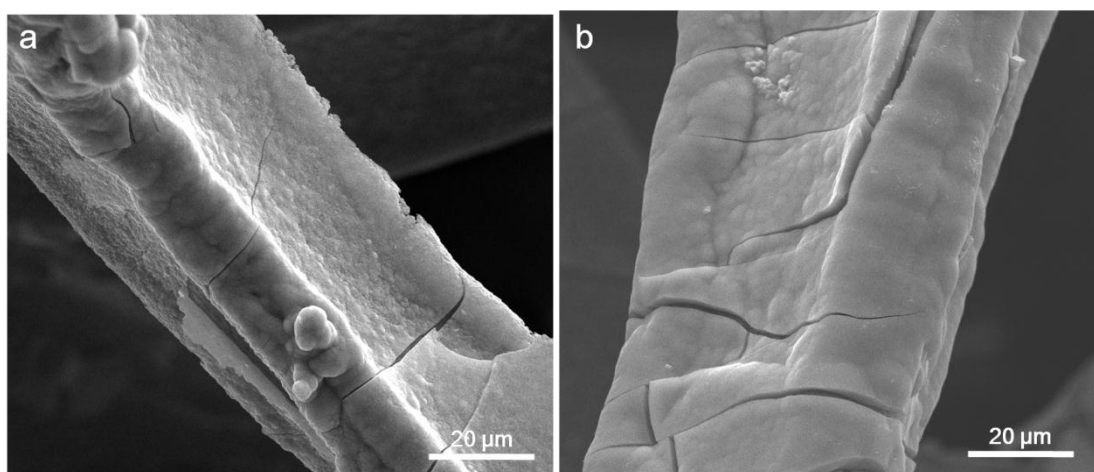


Fig. S12. SEM images of MnO₂/G-gel/NF electrode of ASC (a) before and (b) after 10000 cycles life performance.

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