Supporting Information

A sandwich structure of graphene and nickel oxide with excellent supercapacitive performance

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I. Experimental section

The preparation details of the reference samples are presented as follows.

(a) **GNS membrane:** The powdered GNS was prepared by a low-temperature exfoliation under vacuum [1] and the GNS membrane was assembled by a method that was recently proposed by our group.[2] The assembly procedure is briefly presented as follows (for details see Ref. 2). A GNS suspension was prepared using sodium lignosulphonate (SLS) as the dispersant and a stable mixture suspension of GNS/GO was obtained by homogenously mixing two suspensions where the mass ratio of GNS relative to GO is 9:1. Then, the mixture suspension was heated up to 80 °C for a period in a thermostat water bath, during which a smooth and condensed thin membrane was formed at the liquid-air interface. Such stable membranes were easily taken out and dried at 80 °C for several hr and a free-standing macroscopic GNS membrane was finally obtained.

(b) **Powder-like GNS/NiO hybrid with a randomly aggregated structure:** 100 mg GNS was dispersed in 100 mL aqueous solution under ultra-sonication, where hexadecyltrimethyl ammonium bromide (CTAB) was used as the dispersing agent. Then, 1 mL $Ni(NO_3)_2$ solution was introduced into the dispersed GNS under sonication for 1 hr. Next, ammonia solution ($NH_3 \cdot H_2O$, 25 wt%) was added slowly into the above suspension under stirring until the precipitation appeared. The precipitation was filtered, washed with DI water and dried at 70 °C. Finally, the dried precursor was calcined at 500 °C for 5 hr to obtain the powdered GNS/NiO hybrid with a randomly aggregated structure.

(c) **NiO paticles:** NiO particles were prepared by the same procedure as that for GNS/NiO preparation but in absence of GNS. The details are as follows. 100 mL Ni $(NO_3)_2$ solution (0.1 mol/L) was slowly dried at low temperature (70 °C) for 48 hr under vacuum. Then, the dried powders was thermally treated at 500 °C for 5 hr under high vacuum (<10 Pa) to obtain the powdered NiO.

(d) **RGO membrane:** A GO membrane was obtained by the evaporation of GO hydrosol with the same condition (70 °C, 48 hr) as that for GO/$Ni(NO_3)_2$ hybrid membrane. Then the as-prepared GO membrane was reduced into RGO membrane at 500 °C for 5 h under vacuum (<10 Pa), exactly the same as the reduction of GO/$Ni(NO_3)_2$ membrane.

II. XPS measurment of GNS/NiO sandwich membrane

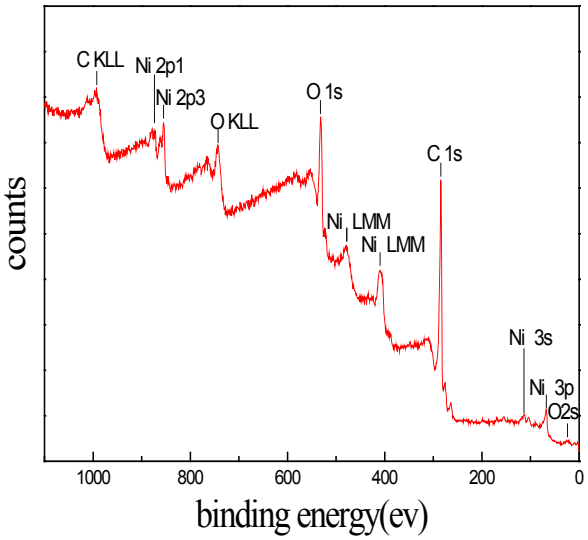

Figure S1. XPS wide-scan spectrum of the as-prepared GNS/NiO sandwich membrane

III. EDS spectrum of a GNS/NiO sandwich membrane

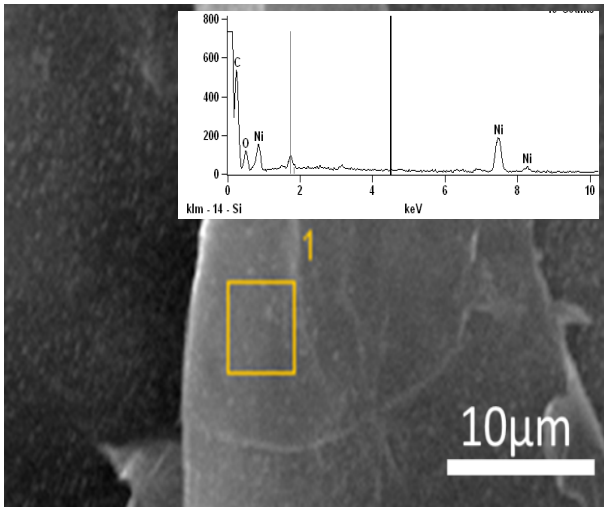

Figure S2. EDS spectrum of the as-prepared GNS/NiO sandwich membrane

No nitrogen was detected in the XPS and EDS measurements for the obtained GNS/NiO sandwich membrane, indicating a full decomposition of $Ni(NO_3)_2$ to NiO.

IV. TG profile of a GNS/NiO sandwich membrane

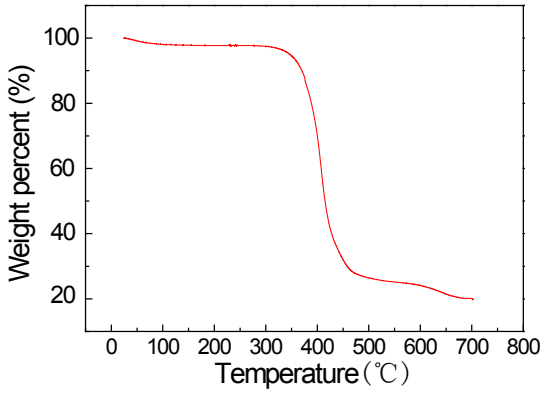

Figure S3. TG profile of the as-prepared GNS/NiO sandwich membrane

V. CV performance of a GNS/NiO sandwich membrane relative to a powdered GNS

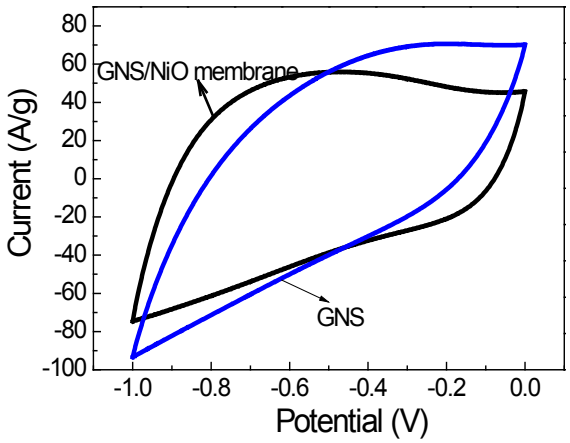

Figure S4. CV profiles of GNS/NiO membrane and GNS at the scan rate of 500 mV/s.

VI. SEM and TEM images of GNS/Co_3O_4 and GNS/Fe_3O_4 sandwich membranes prepared using the same approach

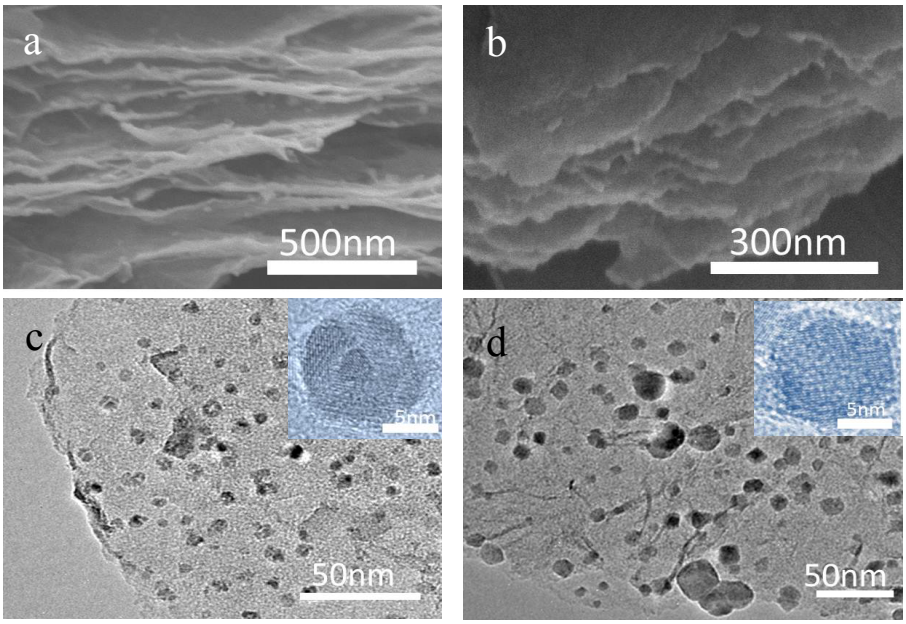

Figure S5. SEM images of GNS/Co_3O_4 (a) and GNS/Fe_3O_4 (b) sandwich membranes; TEM images of GNS/Co_3O_4 (c) and GNS/Fe_3O_4 (d) sandwich membranes. The overlays of (c) and (d) respectively represent the high-resolution TEM images of Co_3O_4 and Fe_3O_4 NPs.

VII. Electrochemistry measurments of as-prepared GNS/Co_3O_4 and GNS/Fe_3O_4 sandwich membrane

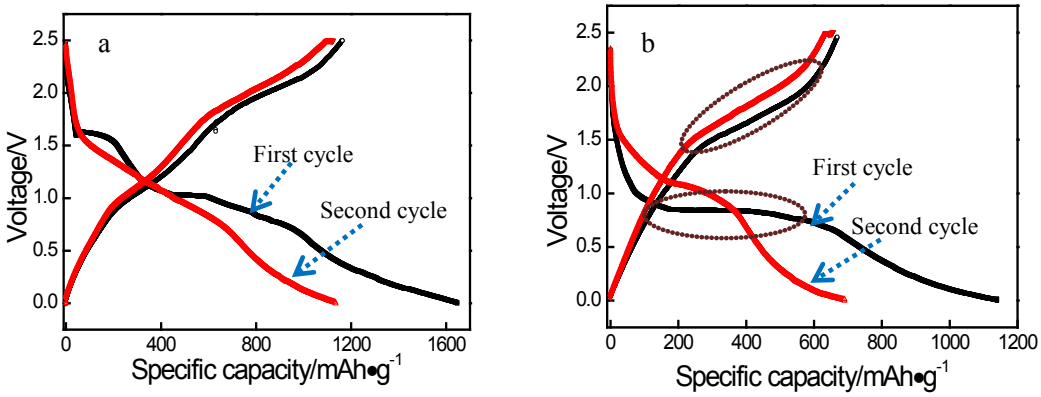

Figure S6. Charge-discharge (current density: 20 mA/g) profiles of sandwich membranes ((a), GNS/Co_3O_4; (b), GNS/Fe_3O_4) that were used as the anodes of Li-ion battery packs. For a larger current density (100 mA/g, not shown here), the discharge capacities of the above membranes are 550 and 480 mAh/g for the first cycle and still remain 490 and 420 mAh/g respectively after 40 cycles.

References:

1. W. Lv, D. M. Tang, Y. B. He, C. H. You, Z. Q. Shi, X. C. Chen, C. M. Chen, P. X. Hou, C. Liu and Q. H. Yang, *ACS Nano*, 2009, **3**, 3730-3736.
2. W. Lv, Z. X. Xia, S. Wu, Y. Tao, F. M. Jin, B. Li, H. Du, Z. P. Zhu, Q. H. Yang and F. Kang, *J. Mater. Chem.*, 2011, **21**, 3359-3364.