Supporting Information

Materials and Methods

Graphene Ink Preparation. As-purchased graphite flakes (Sigma Aldrich) and sodium cholate surfactant (Sigma Aldrich) (0.5 mg/mL) were first mixed in DI water with initial graphite concentration of 3 mg/mL. The mixture was then subject to ultrasonication for 300 min in an ultrasonic bath (Branson 2510). The resulting dispersions were left to sit for ~12 hr allowing unstable aggregation to form and precipitate, and then transferred to vials and centrifuged at 1500 rpm for 60 min (Sorvall RC-5C). After centrifugation, the top 75% of the dispersion in vials was carefully removed by pipit and retained for use as graphene ink solution. Absorption measurements were taken using a Varian Cary 6000i to estimate the concentration of as-prepared ink solutions.

Textile Coating with Graphene. A piece of textile sheet made of randomly intertwined polyester fibers (Cloud 9 dream fleece, Wal-Mart) was dipped into the graphene ink and immediately removed. Due to strong absorption, the ink quickly coated the textile. The 'wet' textile was then placed in the oven for 10 min drying at 120 °C to remove the water. This simple 'dip and dry' process was repeated for a number of times to increase the graphene loading. After each 5 cycles of dipping and drying, the textiles were subject to surfactant removal by washing with abundant DI water and subsequently put into 6 M nitric acid solution for 60 min treatment. These additional steps greatly improve the conductivity of prepared textiles and a factor of ~3 improvement in sheet resistance has been observed for most samples. The mass of coated graphene was obtained by the weight difference before and after graphene coating of the textiles.

MnO_2 Electrodeposition. Typically, a piece of graphene-coated textile was cut with size of 1 cm × 2 cm, and 1 cm × 1 cm was immersed into the aqueous solution with 20 mM $Mn(NO_3)_2$ and 100 mM $NaNO_3$. Electrochemical deposition of MnO_2 nanomaterials was achieved with a three electrode setup, where the conductive graphene-textile was used as the working electrode, platinum electrode (Fisher Scientific) as the counter electrode and Ag/AgCl electrode (Fisher Scientific) as the reference electrode. A small piece of Pt foil (Sigma Aldrich) is used to connect the textile and the crocodile clips to avoid potential side reactions during deposition. A small constant current 100 $\mu A/cm^2$ was applied for a range of deposition time 30~300 min to ensure the conformal coating of MnO_2 nanostructures on graphene-coated textile fibers. After electrodeposition, the textiles were taken out and carefully washed with DI water to remove excessive electrolyte, and then placed on a

sheet of clean paper in vacuum oven at 50 °C to let dry for 2 hr. Overheating of electrodeposited textiles would lead to the cracking of MnO_2 film, especially for thick film resulted from long deposition time. The mass of deposited MnO_2 nanomaterials was again obtained by the weight difference before coating and after drying of the textiles.

Electrochemical Measurement. The electrochemical characterization was carried out using a BioLogic VMP3 potentiostat-galvanostat multichannel equipped with electrochemical impedance spectroscopy (EIS) board. A Ag/AgCl reference electrode (Fisher Scientific) was used in the measurement. Cyclic voltammetries were performed in the potential range of 0 V to 0.85 V vs. Ag/AgCl reference electrode under a sweep rate of 2~100 mV/s.

EC Full Cell Fabrication & Test. The full cells were assembled as coin cells, with as-made graphene/MnO_2–textile as the positive electrode, SWNT-textile as the negative electrode, nickel foam as current collectors, and Whatman filter paper as the separator sandwiched in between two electrodes. The electrolyte added in the cell was 0.5 M Na_2SO_4 aqueous solution with pH=10. Galvanostatic measurements were made using a Biologic VMP3 multichannel system and MTI battery analyzer (Maccor 4300).

Data analysis. In the galvanostatic charging/discharging data, the iR drop from the top cut-off potential and the slope of the discharge curve were used to obtain the power and energy densities, respectively. The power density and energy density were calculated by using the following equations:

$$P = V^2/[4RM] \quad \& \quad E = 0.5CV^2/M,$$

where V is the applied voltage, R is the internal resistance, M is the total mass of active electrode materials used in EC, and C is the measured total capacitance of hybrid EC cell.

The internal resistance was determined from the voltage drop at the beginning of each discharge:

$$R = \Delta V_{iR}/2i,$$

ΔV_{iR} is the voltage drop between the first two points from its top cut-off. This voltage drop is also referred to as the iR drop. i is the current applied.

The total capacitance C of hybrid EC was calculated by using

$$C = i/-[\Delta V/\Delta t] = i/-(\text{slope}),$$

where i is the current applied, $[\Delta V/\Delta t]$ is the slope of the discharge curve after the initial iR drop.

Supporting Figures

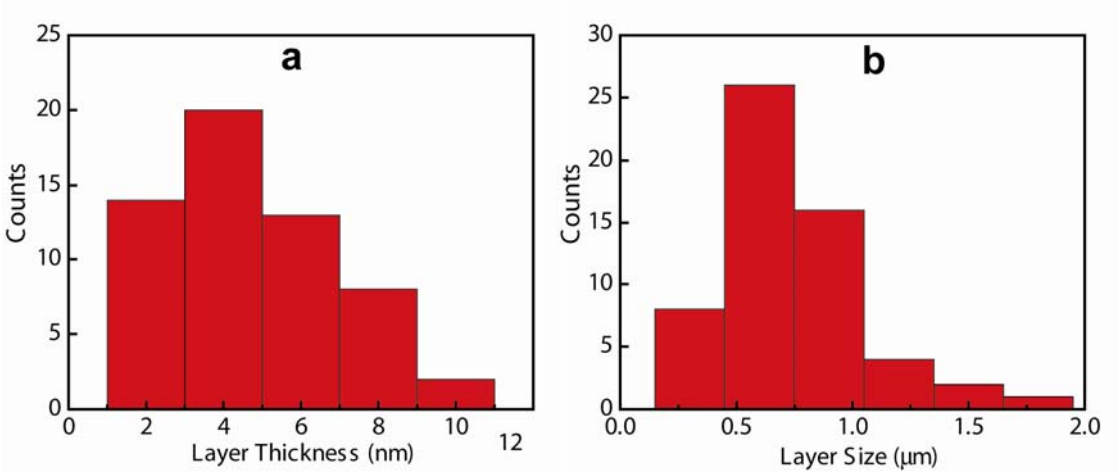


Figure S1. Histograms of AFM analyses on solution-exfoliated graphene nanosheets for (a) layer thickness, and (b) layer size distribution. The samples were prepared with initial graphite concentration of 3 mg/mL, sodium cholate concentration of 0.5 mg/mL, sonication time of 300 min and centrifugation time of 60 min at 1500 rpm.

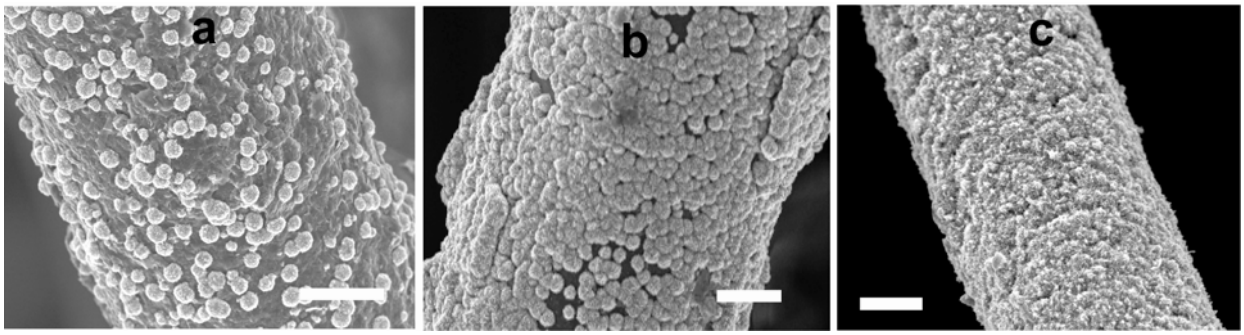


Figure S2. SEM images of electrodeposited MnO_2 nanostructures on graphene-coated textiles with three different deposition times of 30 min (a), 120 min (b), and 240 min (c), respectively. Scale bars: 5 μm for all three images.

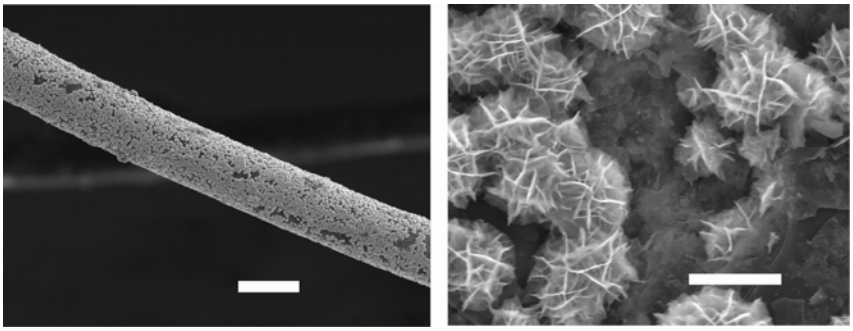


Figure S3. SEM images of a graphene/MnO_2–textile sample after 5000 cycles showing that the MnO_2 was still uniformly coated on textile fibers (left) and the nanoflower MnO_2 structures were well maintained (right). Scale bars are 20 μm in the left image and 1 μm in the right image.