Supplementary Information

Designing Multifunctional Supercapacitor Electrodes Using an Informatics Approach

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S1. Experimental Methods

S1.1 Materials

Kevlar 69 thread was purchased from Thread Exchange. Carboxylic acid functionalized carbon nanotubes were purchased from CheapTubes. Graphite (SP-1) was purchased from Bay Carbon. Dimethyl sulfoxide (DMSO), sodium nitrate ($NaNO_3$), hydrogen peroxide (H_2O_2), and potassium permanganate ($KMnO_4$) were purchased from Sigma Aldrich. Potassium hydroxide (KOH) was purchased from Amresco. Carbon paper was purchased from Alfa Aesar. Microporous poly(propylene) separator (Celgard 3501) was provided by Celgard.

S1.2 Preparation of functionalized CNTs

To prepare functionalized carbon nanotubes, 40 mg of carbon nanotubes were added to a solution of 90 mL H_2SO_4 and 30 mL of HNO_3. The mixture was heated to 60 ^oC and stirred for 4 hours. Then, the mixture was added to 800 mL of deionized water in an ice bath and stirred overnight. Vacuum filtration was used to isolate the CNTs. The CNTs were redispersed in deionized water and purified using dialysis to remove residual acid. Finally, the deionized water was evaporated using a oven at 70 ^oC. The resulting functionalized carbon nanotubes were collected and crushed using a mortar and pestle into a fine powder.

S1.3 Preparation of ANFs

To prepare the ANF dispersion, 1 g of Kevlar 69 thread and 1.5 g of KOH were added to 500 mL of DMSO and stirred for 1 week.[27] The ANF solution was diluted to 0.2 mg mL^{-1} for use in electrode preparation.

S1.4 Preparation of graphene oxide

Graphite oxide was synthesized from graphite using a modified Hummers method.[49] 3 g of graphite powder and 2.5 g of $NaNO_3$ was added to 120 mL of H_2SO_4 while in an ice bath and stirred for 5 hours. Next, 15 g of $KMnO_4$ was slowly added to the mixture over the course of 30 minutes while stirring. An ice bath was used to keep the temperature below 20 ^oC. Then, the mixture was heated to 35 ^oC and stirred for 2 h. The mixture was then added to 250 mL of deionized water. The mixture was stirred for 20 minutes and then added to 700 mL of deionized water. 20 mL of 30 wt % H_2O_2 was then added to the mixture and stirred for 20 minutes. The mixture was washed with 5 wt % HCl solution and filtered using vacuum filtration. The filtered mixture was re-dispersed in deionized water and purified using dialysis. The dispersion was dried at 70 ^oC in an oven. The graphite oxide powder was crushed into a fine powder using a mortar and pestle. The graphite oxide powder was dispersed in deionized water, at 1.0 mg mL^{-1} and exfoliated using ultrasonication to give graphene oxide sheets. Solvent exchange was performed to obtain a GO/DMSO dispersion. Briefly, 100 mL of DMSO was added to a 100 mL solution of GO in water (1.0 mg mL^{-1}) the mixture was sonicated for 1 hour and then rotary evaporation was used to remove the water.

S1.5 Preparation of composite GO/ANF/CNT Electrodes

GO/DMSO (1.0 mg ml^{-1}) and CNT/DMSO (0.2 mg mL^{-1}) dispersions were sonicated for 1 hour. GO/DMSO, CNT/DMSO, and ANF/DMSO (0.2 mg mL^{-1}) were mixed together in desired ratios and stirred for 1 hour. The total amount of material was held constant at 40 mg for every electrode. The mixture was then heated to 80 ^{o}C and deionized water was added (1 mL for every 1 mg of ANF) to reprotonate the ANFs. The mixture was stirred for 1 hour and then allowed to cool to 40-50 ^{o}C. The mixture was then vacuum filtered using a nylon membrane filter (47 mm in diameter, 0.2 μm of pore size). The electrode, while still on the membrane, was washed twice with deionized water and then air-dried overnight. The electrode was then carefully peeled off of the membrane and then dried between two microscope slides in a vacuum oven at 80 ^{o}C for 3 days.

S1.6 Preparation of Composite rGO/ANF/CNT Electrodes

GO/ANF/CNT electrodes were fabricated at varying compositions using flow-directed self-assembly accomplished through vacuum filtration. The electrode was thermally reduced at 200 ^{o}C for 2 hours to reduce the GO sheets into rGO sheets. The compositions presented in the main text represent the weight fractions of GO, ANFs, and CNTs. We assume that there is negligible mass loss upon reduction of GO, such that the weight fractions can be equivalently expressed using rGO. In actuality, there will be some mass loss due to the elimination of oxygen-containing functional groups on the GO sheets, so that the actual rGO content will decrease.

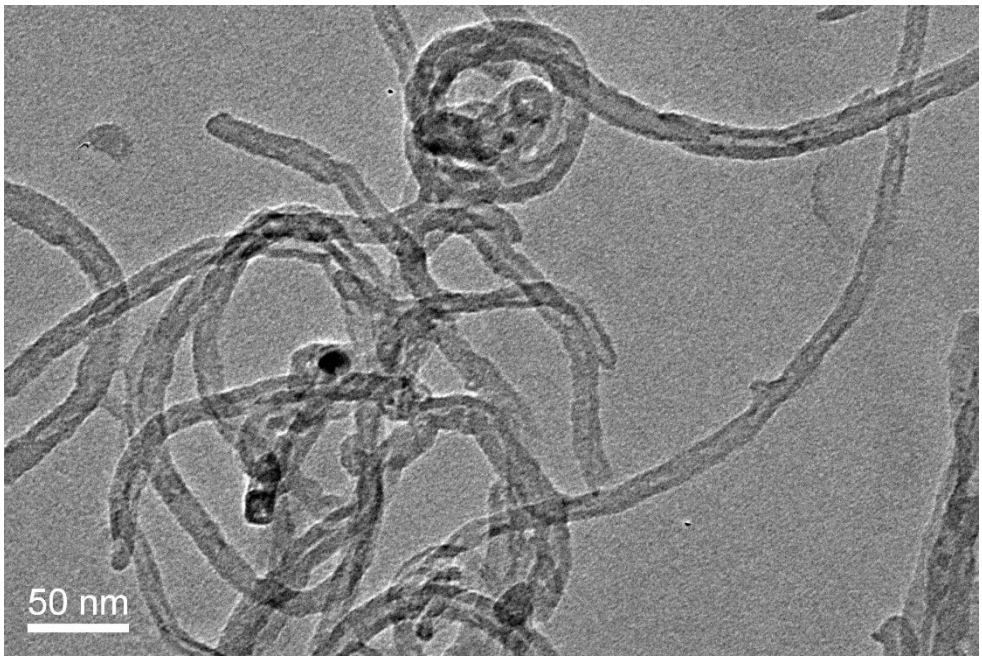

Figure S1. TEM image of CNTs drop cast onto a TEM grid.