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# **Electronic Supplementary Information**

# Shape-Engineerable Composite Fibers and Their Supercapacitor Application

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### Materials and method

#### Materials

Large GO flakes that were stably dispersed in DI water were used for fiber spinning and are called GGO in this paper. The average GO size was  $\sim 37 \mu m$ . The SWNT powder (average diameter; 1.3 to 1.5 nm, thickness  $\sim 20 nm$ , purity 60–70 wt% (90 vol%), catalyst metal; 10 wt%, and graphitic impurities; 20 wt%) were supplied by Hanwha Nanotech (South Korea). SDBS as a surfactant and PVA (Mw: 146,000–186,000; hydrolysis: 99%) were purchased from Sigma-Aldrich (USA). Sulfuric acid liquid electrolyte (1M H<sub>2</sub>SO<sub>4</sub>) was purchased from Daejung Chemical (South Korea).

#### Wet-spun GGO/SWNT/PVA Ribbon

0.3 wt% SWNT was dispersed in DI water with 1 wt% SDBS surfactant using ultrasonication for 1 h. The SWNT/water dispersion was mixed with a dispersion of 0.3 wt% GGO in water by hand shaking. The final concentrations of SWNT, GGO, and SDBS were 0.15, 0.15, and 0.5 wt%, respectively. The GGO/SWNT/SDBS dispersion was injected at an injection rate of 24 mL/h into a 5 wt% PVA coagulation bath. The PVA bath was placed on a stage rotating constantly at 10 rpm during spinning. The GGO/SWNT ribbon gel came up to the surface of the PVA solution just after spinning. The output mass of the GGO/SWNT/PVA was about 860 mg per hour. The GGO/SWNT/PVA ribbon gel was formed and sank to the bottom of the coagulation bath after 18 to 24 h in an oven at 60 °C. The final width of the ribbon gel was 500 to 700  $\mu$ m.

## Characterization

The surface and cross-section morphology of the GGO/SWNT hollow fiber, twisted fiber, and ribbon were obtained using SEM (S4700; Hitachi, Japan). The diffraction intensities from the ribbon before and after annealing were measured using an XRD apparatus (D8 Advance; Bruker, USA). Mechanical properties of hollow fibers, twisted fibers, and ribbons were characterized using a universal testing machine (Instron 5966, USA). Cyclic voltammetry and electrochemical impedance spectroscopy measurements were obtained using electrochemical analyzers (Gamry Instruments, USA).



**Fig S1.** Cross sectional SEM image of hollow graphene/CNT fiber which was annealed at 600 °C for 2 hours. The red circle indicates graphene particle in the fiber.



**Fig S2.** CV curves of rGO/SWNT ribbon fibre measured in three electrode system using Ag/AgCl and Pt mesh as reference and counter electrode, respectively. Liquid electrolyte of 0.1 M Na<sub>2</sub>SO<sub>4</sub> solution is used.