Supporting Information

Ultracompressible, High-Rate Supercapacitors from Graphene-Coated Carbon Nanotube Aerogels

*Evan Wilson and Mohammad F. Islam**

Department of Materials Science & Engineering, Carnegie Mellon University, 5000 Forbes
Avenue, Pittsburgh, Pennsylvania 15213, United States

*Corresponding Author: Tel.: +1 412 268 8999; Fax: +1 412 268 7596; E-mail:
mohammad@cmu.edu.

Figure S1. Structural characterization of the graphene-coated nanotube aerogels. (a) Images of various aerogel shapes. (b) Micrograph from scanning electron microscope of aerogels showed an open porous network structure. (c) Transmission electron microscopy showed nearly all nodes and about 12% of the struts of the network were coated with graphene.

Figure S2. Raman spectra of the pristine and graphene-coated nanotube aerogels. Intensity of each spectrum was normalized to its G-band intensity, which corresponded to the amount of sp^2 -hybridized carbon within the network.^[1] The ratio between the G-band and the D-band intensities, labeled on the plot as G and D respectively, served as a measure of the relative amount of sp^3 -hybridized defects on the wall and was used to quantify damage to the nanotubes during graphene coating. Note, D-band intensity corresponded to sp^3 -hybridized carbon in the aerogels. It was expected that if nanotubes underwent damage, the intensity ratio of G-band to D-band should decrease.^[2] For the graphene coated nanotube aerogels, the intensity ratio of the G-band to the D-band was largely unchanged compared to pristine nanotube aerogels, indicating little to no damage during the graphene-coating as well as thorough reduction of the deposited glucose. Likewise, the radial breathing modes (RBMs) were intact after the graphene coating and did not display a significant shift or decrease in intensity, which indicated that the nanotubes were not significantly functionalized or bundled. We attribute the slight change in peak locations for the RBMs from pristine to graphene-coated nanotube aerogels to interactions between the nanotubes and graphene.^[3]

Figure S3. Adsorption isotherms and pore characteristics of the aerogels. (a) N_2 adsorption/desorption isotherms of the aerogels of mass density of 9 mg/mL at 77 K is shown by plotting experimentally measured specific adsorbed volume (i.e., adsorbed volume divided by mass of the sample) versus relative pressure P/P_0 , where equilibrium pressure and saturation pressure of the adsorbate at the adsorption temperature of 77 K are denoted as P and P_0 , respectively. (b) Pore diameter distribution (dV/dr) versus pore diameter ($2r$) of the aerogels, calculated from the desorption branch of the isotherm by both Barrett-Joyner-Halenda (BJH) and density functional theory (DFT) methods.

Figure S4. A schematic of the cells that were used for the compression tests. (a) Cross-section of the cell showing multiple shims that we used to vary the thickness of the cells, the stainless steel current collectors that were in intimate contact with the graphene-coated nanotube aerogel electrodes, and a porous separator maintaining electrical isolation between the electrodes. (b) Cells with compressed electrode were constructed with a single spacer shim. The unoccupied volume of the cells allowed electrolyte to flow out of the electrode during compression and be taken up during release. Over the course of the measurements, we did not need to add any electrolyte to the cells.

Figure S5. Dependence of gravimetric capacitance for cells with aqueous electrolyte on thickness of the electrodes. The gravimetric capacitance of a cell with 1.5 mm thick electrodes was comparable to that of a cell with electrodes nearly an order of magnitude thinner at low scan rates. Furthermore, the capacitance of the cells with 1.5 mm electrodes was similar when compressed to 150 μm to that of cells with similar thickness electrodes, indicating strain-invariance of the ionic transport through the electrodes.