# Supporting Information

# Templated 3D ultrathin CVD graphite networks with controllable geometry: synthesis and application as supercapacitor electrodes

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## Experimental Details

*Sample Preparation*

Silicon (100) substrates, 6" in diameter, are cleaned via a rinse in piranha solution (50:1 $H_2SO_4$:$H_2O_2$, 120 °C), followed sequentially by rinses with deionized (DI) water, concentrated HF solution, and DI water. A 100 nm layer of Ni, with a 5 nm Ti adhesion layer, is deposited onto the Si wafer in an e-beam evaporator (Thermionics). This metal layer serves as the seed for subsequent Ni electrodeposition. The wafer is diced into 1.5 x 1 $cm^2$ chips and cleaned with ultrasonication in acetone followed by sequential rinses with isopropyl alcohol and DI water.

*Microsphere Deposition*

Suspensions of polystyrene spheres of 1 μm and 0.5 μm diameters (Thermo Scientific) are diluted to 1 wt% colloidal suspensions in a 1:1 by weight ethanol to DI water ratio. The sample is partially immersed into a beaker filled with water and the diluted suspension is deposited onto the water surface via micropipette. A drop of 3% sodium dodecyl sulfate solution is used to order the spheres on the surface into a close-packed array. The sample is slowly withdrawn from the beaker, leaving a monolayer of spheres on the Ni surface, then dried and annealed for 30 seconds at 100 °C on a hot plate. The process is repeated to add additional layers of spheres. After the desired number of layers is deposited, the sample is annealed for 5 minutes at 100 °C on a hot plate in order to partially sinter the spheres together.

*Electrodeposition of Ni*

A high throwing power Ni sulfate bath (composed of 0.775 M $NiSO_4$, 1.173 M $NiCl_2$, 2.534 M $NaSO_4$, and 0.809 M boric acid) is used for the electrodeposition. [1] The deposition is performed with stirring at room temperature, using a deposition current of 3 $mA/cm^2$ and a Ni wire counter

electrode. After deposition of the desired amount of Ni, the sample is immersed in tetrahydrofuran (Sigma-Aldrich) overnight to dissolve the polystyrene spheres.

*Ultrathin graphite growth*

After loading the sample in the center of a hot-wall CVD tube furnace (Thermo Scientific Lindberg Blue M), a crucible with 1 mL benzene (Sigma-Aldrich) is placed near the inlet. The crucible is covered with aluminum foil with a small hole (diameter ~ 1 mm). After flowing 200 sccm of Ar (Praxair) for 10 min at ambient pressure, $H_2$ gas (Praxair) is introduced at a 65 sccm flow rate, and the temperature is raised to 600 °C at ~ 40 °C/min under a combined $H_2$ and Ar flow. Once the furnace reaches the desired temperature, the furnace is turned off, and the sample is then allowed to cool at a rate of ~ 25 °C/min in the $H_2$ + Ar flow to yield a multilayer graphene which has sufficient mechanical stability for a free-standing foam, without polymer supports required for transfer. [2] After deposition, the Ni is etched overnight in concentrated 12 M HCl. Upon etching, the freestanding carbon foam floats to the surface, where it is scooped up by another substrate (polycarbonate sheets in this case).

*Calculation of energy and power density*

The reported energy and power density are calculated from the cyclic voltammetry data at a scan rate of 50 mV/s and 10 V/s respectively using the following formulae:

$$E = \frac{1}{2} C V^2$$

$$P = \frac{E}{\Delta t}$$

where $E$ is the energy density, $P$ is the power density, $C$ is the specific capacitance (either areal or volumetric), $V$ is the voltage window scanned (0.8 V), and $\Delta t$ is the discharge time.

References:

(1) Di Bari, G. A., In *Modern Electroplating;* Schlesinger, M., Paunovic, M., Eds.; John Wiley & Sons, Inc.: Hoboken, NJ, 2010; Chapter 3, pp 79-114.

(2) Ji, H.; Zhang, L.; Pettes, M. T.; Li, H.; Chen, S.; Shi, L.; Piner, R.; Ruoff, R. S. Ultrathin Graphite Foam: a Three-Dimensional Conductive Network for Battery Electrodes. *Nano Letters* **2012,** *12,* 2446-2451.