## **Supporting information**

### Materials.

The SBA-15-supported carbon nanomembrane (denoted SS-CNM-2) was prepared according to our recently published procedure [1]. SS-CNM-1, SS-CNM-3 and SS-CNM-4 were prepared according to a similar procedure with MCM-41 [2] and pore-expanded SBA-15s [3,4] as templates, respectively.

#### Characterizations

The morphology of the samples was investigated by using a transmission electron microscope (TEM, JEOL, JEM-2010) operated at 200 kV. X-ray diffraction (XRD) patterns were conducted on a RIGAKU D/MAX 2550 VB/PC X-ray diffractometer (Cu Kα radiation generated at 40 kV and 450 mA). TGA were conducted on a TG/WRT-2P in air. Nitrogen adsorption/desorption isotherms were measured with an adsorption apparatus on an ASAP2010 instrument. The surface area of the samples was determined from the Brunauer-Emmett-Teller (BET) equation and pore volume from the desorption branches of the isotherms with use of the Barrett-Joyner-Halanda (BJH) method.

# **Theoretical Calculations**

Cylindrical pores are generally used as a theoretical model for porous electrode materials. By assuming that the pores are cylindrical, solvated ions enter pores and approach the pore walls to form electric double-cylinder capacitors. The capacitance of this double-cylinder capacitor is thus directed by the number of solvated ions that are hosted by the cylinder electrode in the compact layer. Although the diffuse layer may make a small contribution to the capacitance (especially when the diameter of the cylinder is much larger than the diameter of solvated ions), it is typically elided as the compact layer encompasses much of the potential drop.

Depending on the diameter (or radius) ratio (R/r) of the cylindrical electrode and spherical solvated ion, the hosting capacity (n) of a cylinder for spheres is discussed in two scenarios: when R/r is smaller than 2, the solvated ions line up in the cylinder to form a wire-in-cylinder capacitor, and a zigzagging wire forms for maximum hosting capacity; when R/r is larger than 2, a double-cylinder capacitor forms, and the hosting capacity can be calculated according to the included angle that each solvated ion occupies in the compact layer.

The capacitance of a porous electrode material is calculated with the following equation, taking the fractal dimension (D) [1] into account.

$$C = K n / (R/r)^{(D-1)}$$

The fitting curves of an ideal 2D tube array, ideal 3D porous object, and porous (2.5D) material are presented in Figure S2.



**Figure S1**. Calculation of the theoretical hosting capacity of a cylindrical electrode for spherical ions, simplified as circles in a rectangle (R/r < 2) and circles in a circle (R/r > 2).



Figure S2. Schematic depiction of the relation between pore size and theoretical specific capacitance of an EDLC. Plots for ideal three-dimensional (D = 3) porous material and an array of ideal two-dimensional (D = 2) tubes are shown in red and black, respectively. A porous object (blue line) with a fractal dimension [5] D = 2.5 is used to mimic the mesoporous carbon materials typically used in an EDLC.



Figure S3 Cycling stability plots for SS-CNM-2 in aqueous electrolyte solutions.



Figure S4 Drawing of ideal EDLC electrode material, Honeycomb⊂Honeycomb.

## References

 Yang, X., et al., Synthesis of Ultrathin Mesoporous Carbon through Bergman Cyclization of Enediyne Self-Assembled Monolayers in SBA-15. Langmuir, 2010.
26(13): p. 11244-11248.

[2] Cai, Q., et al., Dilute Solution Routes to Various Controllable Morphologies of MCM-41 Silica with a Basic Medium. Chemistry of Materials, 2001. 13(2): p. 258-263.

[3] Ru, H.Q., et al., A facile and versatile partitioned cooperative self-assembly process to prepare SBA-15s with larger mesopores, high microporosity and tunable particle sizes. Journal of Material Chemistry, 2011. 21(32): p. 12059-12067.

[4] Fulvio, P.F., S. Pikus, and M. Jaroniec, Tailoring properties of SBA-15 materials by controlling conditions of hydrothermal synthesis. Journal of Materials Chemistry, 2005. 15(47): p. 5049-5053.

[5] Avnir, D. and M. Jaroniec, An isotherm equation for adsorption on fractal surfaces of heterogeneous porous materials. Langmuir, 1989. 5(6): p. 1431-1433.