## **Supplementary Information**

## Carbon nanotube 3D current collectors for lightweight, high-performance and low-cost supercapacitor electrodes

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**Fig. S1** Formation of the AC-FWCNT hybrid electrode through vacuum filtration on PTFE membrane (a). Schematic of the Ti-mesh line contact formation. The arrows indicate the vertical/parallel electric conduction and vertical ionic diffusion through the hybrid film.



**Fig. S2** N<sub>2</sub> sorption isotherms at 77 K (a) and pore size distribution (BJH plot) (b) of the FWCNT, AC and AC-FWCNT films. The adsorption isotherm and pore diameter distribution of the AC-FWCNT hybrid electrode are close to that of the AC electrode, with a slight increase in mesopore content. The surface area of the AC-FWCNT film is 1783 m<sup>2</sup>/g, which is consistent with the individual specific surface area values for FWCNTs (428 m<sup>2</sup>/g) and the AC, YP80-F (2034 m<sup>2</sup>/g).



**Fig. S3** Change of sheet conductivity with CNT weight fraction. The values (taken from this work and from previous studies) are for self-supporting electrodes combining CNTs and AC for supercapacitor applications. The type and source of CNTs is indicated, since these are important parameters which determine their conductivity. The AC-FWCNT values correspond to films ~200  $\mu$ m thick and with densities of 0.25–0.30 g/cm<sup>3</sup>.



**Fig. S4** Rate capabilities of conventional electrodes of AC (90 wt%)-AB (5 wt%)-PTFE (5 wt%) (the same data as Fig. 4d) and AC (85 wt%)-AB (10 wt%)-PTFE (5 wt%) with Ti mesh in full-, line- and point-contact configurations. The AC (85 wt%)-AB (10 wt%)-PTFE (5 wt%) electrode supported on a Ti foil and sandwiched with Ti meshes was also examined.

Compared with the AC (90 wt%)-AB (5 wt%)-PTFE (5 wt%) electrode (Fig. 4d), the rate performance slightly improved owing to the increased content of the conductive fillers (i.e. AB) while the capacitance at low rates slightly decreased due to the decreased content of the capacitive particles (i.e. AC). The Ti foil did not enhance the rate performance of the electrode sandwiched with Ti meshes in full contact. The electrode on Ti foil was prepared by first making the AC-AB-PTFE paste in the procedure in Experimental, and then pressing the Ti-mesh/AC-AB-PTFE paste/Ti foil/Ti-mesh stack at 10 MPa. Polyvinylidene difluoride (PVDF) was also examined as binder in place of PTFE but yielded poorer results.

These results clearly show the need for a full-contact metallic collector in the conventional sample, and highlight the role of the FWCNT matrix.



**Fig. S5** Nyquist plots for the AC (90 wt%), AB (5 wt%), PTFE (5 wt%) electrodes with Ti mesh in full-, line- and point-contact configurations (a); high-frequency region (b); equivalent electrical circuit showing solution (R1) internal (ESR) and diffusion ( $R_M$ ) resistances.

## References

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