

## Support information:

### Experimental

*Oxidation of multi-walled carbon nanotubes (MWCNT):* 10g MWCNT was stirred in a 200ml mixture of nitric acid and sulfuric acid (3:1 by volume) at 140 °C for an hour. The sample was diluted with DI water and then the oxidized CNTs were collected by centrifuge centrifuged and finally dried.

*Preparation of MFC/MWCNT supercapacitor electrodes:* A slurry was made with 40wt% MWCNT, 40wt% polyethylene oxide (PEO, Mw=3400), 6.7wt% lithium chloride, 10wt% MFC in water. The slurry was stirred in room temperature for an hour and then sonicated for 10 mins. The slurry was then poured into an aluminum disk for water evaporation.

*Preparation of MFC-based separator:* A slurry was made with 70wt% PEO (Mw=3400), 12wt% lithium chloride and 18wt% MFC. The slurry was stirred in room temperature for an hour and poured into an aluminum disk to form sheets.

*Sample Characterizations:* The surface morphology of samples was examined on a LEO 1530 scanning electron microscopy with an acceleration voltage of 3kV. For the electrochemical measurement, the cyclic voltammetry curve was obtained by a two-electrode system using a DS345 30 MHz (Stanford Research Systems) as the voltage source and a SR 570 (Stanford Research Systems) as the current recorder. The galvanostat charging-discharging curve and nyquist plot was obtained by Princeton potential stat. The conductivity of the electrode and separator sheets was measured by a method of Volt-Ampere. The I-V curve was obtained by the Standford Research System. The length and width of the sheets were measured by vernier caliper and thickness was measured by Mitutoyo Caliper. The specific area and pore volume distribution of the electrode sheets were measured by BET (Quantachrome Quadrasorb SI). The samples were first degassed using Flovac Degasser for 24 hours at 50°C under vacuum and then

measured by BET under high purity N<sub>2</sub> at 77k maintained by liquid nitrogen. The surface area and pore size distribution were determined using the Multi point BET model and DFT model respectively. The mechanical properties were measured by Instron 5567 universal testing. Samples were dried at 105 °C to a constant weight and were cut into required sizes and shapes before measurement. The tests were done at ambient condition at a cross-head speed of 5 mm/min and a gauge length of 20 mm.

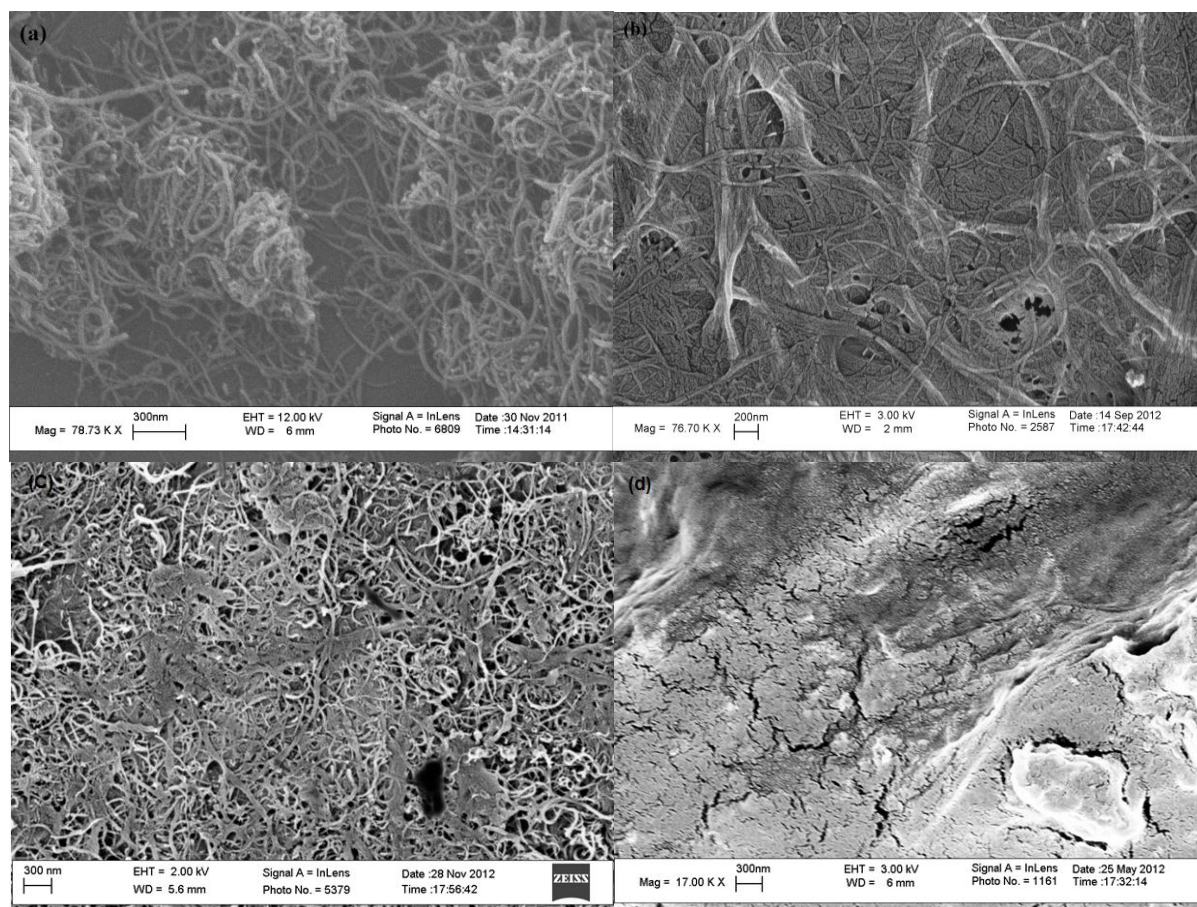


Figure S1. SEM image of a) Multi-walled carbon nanotubes(MWCNT) network; b) Microfibrillated cellulose(MFC).c) MWCNT MFC(80:20 wt%) sheets. d) complexation of PEO and LiCl.

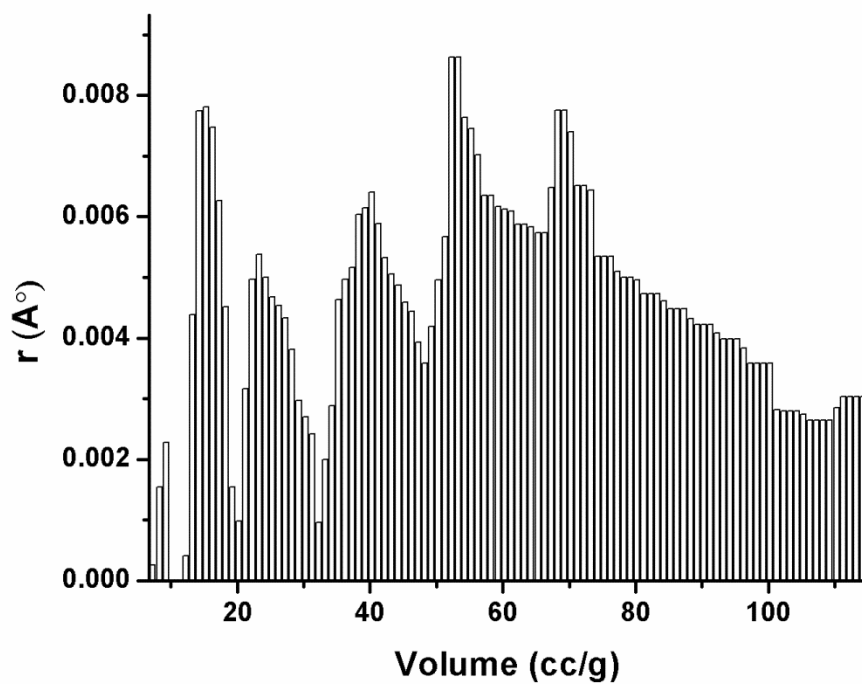


Figure S2. The pore volume distribution in term of pore volume  $V(\text{cc/g})$  v.s. half pore width  $r(\text{\AA})$  of electrode sheet(80wt% MWCNT and 20% MFC).