## Supporting Information Rational Design of Polyaniline/MnO<sub>2</sub>/Carbon Cloth Ternary Hybrids as Electrodes for Supercapacitors

Xin Zhao<sup>1</sup>, Chaoyi Chen<sup>1</sup>, Zilong Huang<sup>1</sup>, Lei Jin<sup>1</sup>, Junxian Zhang<sup>1</sup>, Yingzhi Li<sup>1</sup>, Lili Zhang<sup>2</sup>\*, and Qinghua Zhang<sup>1</sup>\*

<sup>1</sup>College of Material Science & Engineering, State Key Laboratory for Modification of Chemical Fibers and Polymer Materials, Donghua University, Shanghai 201620, China

<sup>2</sup> Institute of Chemical and Engineering Sciences, A\*STAR, 1 Pesek Road, Jurong Island 627833, Singapore

\*Corresponding author. Email: <u>zhang\_lili@ices.a-star.edu.sg</u>, <u>qhzhang@dhu.edu.cn</u>



**Figure S1.** SEM images of original CC and m-CC and their hydrophilic tests (inset shows the contact angle).



Figure S2. SEM images of m-CC after being etched in 1 M  $H_2SO_4$ .



Figure S3. SEM images of  $MnO_2@m-CC$  after being etched by 1 M  $H_2SO_4$ .



Figure S4. TEM images of (a) MnO<sub>2</sub>@PANI@m-CC and (b) PANI@MnO<sub>2</sub>@m-CC



Figure S5. SEM image of the cross-section part of PANI@MnO<sub>2</sub>@m-CC



Figure S6. Raman spectra of  $H_2SO_4$  etched  $MnO_2@m-CC$  and original  $MnO_2@m-CC$ 



**Figure S7.** (a) Survey XPS spectrum of m-CC, PANI@MnO<sub>2</sub>@m-CC, MnO<sub>2</sub>@PANI@m-CC and N 1s spectra of (b) m-CC, (c) PANI@MnO<sub>2</sub>@m-CC and (d) MnO<sub>2</sub>@PANI@m-CC





Figure S9. CV curves of MnO<sub>2</sub>@PANI@ m-CC at various scan rates.



**Figure S10.** Galvanostatic charge/discharge curves of (a) PANI@MnO<sub>2</sub>@m-CC and (b) MnO<sub>2</sub>@PANI@ m-CC at different current densities



Figure S11. Gravimetric specific capacitance vs. current densities of as-prepared samples.



Figure S12. CV curves at 20 mV/s of PANI@MnO<sub>2</sub>@m-CC electrode at flat and bending state



**Figure S13.** Three-electrode electrochemical measurements of a-MEGO@m-CC: (a) CV curves at various scan rates, (b)Galvanostatic charge/discharge curves at different current densities, (c) plot of areal capacitance vs. current density and (d) Nyquist plot from EIS measurement.



**Figure S14.** Comparison of the CV curves of a-MEGO@m-CC and PANI@MnO<sub>2</sub>@m-CC at different potential window in three electrode systems.



**Figure S15.** (a) Galvonostatic charge/discharge curves at a current density of 0.2 mA/cm2 with different potential windows, (b) polts of gravimetric specific capacitance vs. current density, (c) Nyquist plots and (d) cycling stability curves of assembled PANI@MnO<sub>2</sub>@m-CC//a-MEGO@m-CC ASC device.

Samples (1×1 cm <sup>2</sup> )				Mass	s (mg)			
m-CC	9.95	9.51	9.48	9.63	9.87	10.02	9.62	9.90
MnO <sub>2</sub> @m-CC	10.15	9.73	9.70	9.84	-	-	-	-
PANI@m-CC	-	-	-	-	10.22	10.40	9.98	10.25
PANI@MnO2@m-CC	10.50	10.11	10.05	10.21	-	-	-	-
MnO <sub>2</sub> @PANI@m-CC	-	-	-	-	10.43	10.63	10.20	10.47

 Table S1. The total mass of each samples obtained in our work.

Table S2.	Calculated area	al and gravim	etric specific	capacitance o	f a-MEGO@1	m-CC
and PAN	I@MnO <sub>2</sub> @m-C	CC at different	current densi	ties in a three	electrode syst	em.

Samplas	Canacitanaa	Current density (mA/cm <sup>2</sup> )						
Samples	Capacitance	0.2	0.4	0.6	ity (mA/cr 0.8 140 144.4 278.6 488.8	1.0	2.0	
a-MEGO@m-CC	$C_{\rm areal} ({\rm mF/cm^2})$	239.3	158.8	141.2	140	132	132.5	
(0.97 mg/cm <sup>2</sup> )	$C_{\rm m}({\rm F/g})$	246.6	163.7	145.6	144.4	136.1	136.6	
PANI@MnO <sub>2</sub> @m-	$C_{\text{areal}}(\text{mF/cm}^2)$	421.6	337.1	297	278.6	264.1	251.3	
CC (0.57 mg/cm <sup>2</sup> )	$C_{\rm m}({\rm F/g})$	739.6	591.4	521.1	488.8	463.3	440.9	