## Supporting information

Jian Zhi<sup>a,b\*</sup>, Oliver Reiser<sup>a</sup>, Youfu Wang<sup>b</sup>, Aiguo Hu<sup>b</sup>.

<sup>a</sup>Institute of Organic Chemistry, University of Regensburg, Universitätsstr.31,93053

Regensburg, Germany

<sup>b</sup>Shanghai Key Laboratory of Advanced Polymeric Materials, School of Materials Science

and Engineering, East China University of Science and Technology, Shanghai 200237,

China

Email: jian.zhi@outlook.com

## **Capacitance calculation**

If the capacitances of the two electrodes, i.e. positive and negative, can be expressed as  $C_p$  and  $C_n$ , respectively, the overall capacitance ( $C_T$ ) of the entire cell can be expressed as eqn (1):

$$\frac{1}{C_T} = \frac{1}{C_p} + \frac{1}{C_n}$$
(1)

In a symmetrical supercapacitor,  $C_p=C_n=C_0$ , where  $C_0$  represents the per-electrode capacitance and in this study represents the capacitance of one electrode. So the relationship between  $C_T$  and  $C_0$  should be as eqn (2):

$$C_0 = 2C_T = \frac{2It}{V} \tag{2}$$

where I, t and V are charged current, t is the discharge time, V is the voltage drop upon discharging (excluding IR drop).

As a result, the per-electrode areal capacitance  $(C_{A0})$  and volumetric capacitance  $(C_{v0})$  of the device is shown in eqn (3) and eqn (4):

$$C_{A0} = \frac{C_0}{A_0} = \frac{2It}{A_0} = \frac{4It}{A_T V} = 4C_{A-cell}$$
(3)
$$C_{v0} = \frac{C_0}{v_0} = \frac{2I(t - t_{OMC})}{v_0 V} = \frac{4I(t - t_{OMC})}{v_T V} = 4C_{v-cell}$$
(4)

Where  $A_0$  is the surface area of in one electrode, which is approximately considered to be a cylinder.  $A_T$  represents the total mass the whole cell, in which  $A_T=2A_0$ .  $C_{A-cell}$  is the areal capacity of the whole cell.  $V_0$  is the volume of one electrode the total volume of the device  $v_t=2v_0$ .  $C_{v-cell}$  is the volumetric capacitance of the whole device.

The corresponding volumetric energy and power density are calculated through eqn (5) and

(6):

$$E_{v} = \frac{1}{2}C_{v-cell}V^{2} = \frac{1}{8}C_{v0}V^{2}$$
(5)
$$P_{v} = \frac{E_{v}}{t}$$
(6)



Figure S1. Digital image of a vial containing CVD gr/OMC dispersions stabilized by P123 surfactants.



Figure S2. Pore size distribution of CVD gr/OMC below 2 nm calculated from the NLDFT model



Figure S3. XRD pattern of CVD gr after Ni etching.



Figure S4. Absorbance at 660 nm measured for CVD gr/OMC dispersions as a function of sonication time.



Figure S5. The relationship between line resistance and mass loading of CVD gr/OMC.



Figure S6. Ultimate tensile strength of various carbon coated cotton threads.



Figure S7. XRD patterns of MnO<sub>2</sub> spheres.



**Figure S8**. Bulk heterojunction (BJH) pore distribution spectra of the CT-CVD gr/OMC-MnO<sub>2</sub> composite electrode.



Figure S9. The relationship between surface area (a)/pore volume (b) and mass loading of MnO<sub>2</sub> nanoparticles on CT-CVD gr/OMC electrode.



Figure S10. Coulombic efficiency of CT-CVD gr/OMC-MnO<sub>2</sub> supercapacitor as a function of cycling number at a current density of 13.76 mA cm<sup>-2</sup>.



Figure S11. (a) Specific capacitance as a function of scan rate (inverse square root) for CT-CVD gr/OMC-MnO<sub>2</sub> electrode. (b) specific (inverse) capacitance as a function of scan rate (square root) for CT-CVD gr/OMC-MnO<sub>2</sub> electrode.



Figure S12. Bar graph of mass normalized capacitance of MnO2 in various thread-like supercapacitors with Faradaic insertion capacity (red) and Faradaic pseudocapacitive charging (blue) derived from Trasatti's method.



Figure S13. EIS spectra of CT-CVD gr/OMC supercapacitor before and after 3000 cycles.



**Figure S14 (a,b).** EIS data of CT-CVD gr/OMC and CT-CVD gr/OMC- MnO<sub>2</sub> composites and the equivalent circuit diagram used for the fitting.



Figure S15. SEM image of TiO<sub>2</sub> nanowires



Figure S16. (a) Highlights of the response speed under 350 nm illumination. (b)

Highlights of the recovery speed under 350 nm illumination.

Sample	BET-surface area (m <sup>2</sup> g <sup>-1</sup> )	Mesopore volume (cm <sup>3</sup> g <sup>-1</sup> )	Micropore volume (cm <sup>3</sup> g <sup>-1</sup> )	Micro/meso- porosity
CVD-gr	91	0.09	0	-
OMC	236	0.5	0.9	1.8
CVD-gr/OMC	328	0.4	1.1	2.8

Table S1. Structural characterization data for CVD gr, OMC and CVD gr/OMC

 Table S2. Comparison of electrochemical performance of yarn/fiber-based supercapacitors.

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Re	f. Materials	Electrolyte	Voltage	Pv	Ev
			window	mW cm <sup>-3</sup>	mWh cm <sup>-3</sup>
73	B PPy/ MnO <sub>2</sub> /rGO	PVA/ H <sub>3</sub> PO <sub>4</sub>	0-0.8	16	1.1
16	5 ZnO/ MnO <sub>2</sub>	PVA/LiCl	0-0.8	2.4	0.04
74	$TiO_2/MnO_2$	PVA/LiCl	0-0.8	230	0.3
63	MnO <sub>2</sub> /Carbon fiber	PVA/H <sub>3</sub> PO <sub>4</sub>	0-0.8	400	0.22
75	5 PPy	PVA/ H <sub>3</sub> PO <sub>4</sub>	0-0.8	270	1
1.	5 PEDOT/CNT	PVA/ H <sub>3</sub> PO <sub>4</sub>	0-0.8	38000	1.1
76	WO <sub>3-x</sub> /MoO <sub>3-x</sub>	PVA/ H <sub>3</sub> PO <sub>4</sub>	0-1.9	730	1.9
71	V Nickel fiber/Co <sub>3</sub> O <sub>4</sub> nanowire	PVA/KOH	0-1.5	1470	0.62
78	NiCo <sub>2</sub> O <sub>4</sub>	PVA/KOH	0-1	17000	1.44
64	MnO <sub>2</sub> /graphene/carbon fiber	PAAK/KCl	0-1.6	200	0.9
14	CuO/AuPd/ MnO <sub>2</sub>	PVA/KOH	0-0.8	413	0.55
Th	is CVD gr/OMC- $MnO_2$	PVA-BMIMCl-	0-1.5	300	2.7
w0		L12504			1