Supplementary Information

Flexible, highly conductive, and free-standing reduced graphene oxide/polypyrrole/cellulose hybrid papers for supercapacitor electrodes

Caichao Wan, Yue Jiao & Jian Li*

Material Science and Engineering College, Northeast Forestry University, Harbin 150040, China.

**Correspondence and requests for materials should be addressed to J. L. (email: lijiangroup@163.com)*

Detailed procedures of stiffness tests

The stiffness was measured by a Taber stiffness tester (150-E, Taber Industries,) according to the standard Tappi T 489. This method is used to determine the bending moment required to deflect the free end of a 38 mm wide vertically clamped specimen 15° from its center line. The stiffness (i.e., resistance to bending) was calculated from the bending moment, which was read from the instrument scale. The test procedures can be described as follows:

(1) Place a conditioned test specimen (38.1 ± 0.3 mm wide by 70 ± 1 mm long) in the vise with one end approximately level with its top edge and the other end between the rollers.

(2) With the two clamping screws of the vise, align the specimen with the center line of the pendulum.

(3) Turn each of the screws for adjusting the rollers so that they just contact the specimen, then after taking up the backlash in one screw, back off one-quarter turn to give a distance between rollers of 0.33 ± 0.03 mm greater than the thickness of the specimen.

(4) Switch on the motor to rotate the loading disk to the left and thus deflect the specimen until the engraved mark on the pendulum is aligned with the 15° mark on the loading disk. Stop the motor, record the scale reading on the fixed annular disk and immediately return the loading disk to zero. Take a similar reading by deflecting the specimen to the right. The stiffness of the specimen is the average of the two readings multiplied by the factor required for the instrument range weight used.

Table S1. Frequencies (cm^{-1}) of the main signals of the cellulose paper.

Absorption band (cm^{-1})	Assignment
3356	O–H stretching
2904	C–H stretching
1639	bending mode of the absorbed water
1434	CH_2 symmetric bending
1369	O–H bending
1319	C–O symmetric stretching
1161	C–O anti-symmetric stretching
1111	C–OH skeletal vibration
1057	C–O–C pyranose ring skeletal vibration
899	C–H stretching out of plane of aromatic ring

Table S2. Comparison of areal capacitances of RPC papers with other recently reported graphene/PPy composites.

Electrode	Synthesis method	Areal capacitance (F cm^{-2})	Electrolyte	Ref.
Graphene/PPy-deposited titanium metal substrate	Electrodeposition and electropolymerization	0.151 (10 mV s^{-1})	0.1 M $LiClO_4$	[59]
PPy/GO-deposited fluorine-doped tin oxide	Electrochemical codeposition	0.152 (10 mV s^{-1})	1 M KCl	[60]
RGO–PPy membranes	Vacuum filtration	0.175 (10 mV s^{-1})	2 M KCl	[61]
Graphene/PPy composite fibers	Wet-spinning	0.115 (0.2 mA cm^{-2})	H_2SO_4/PVA[a]	[62]
Graphene–PPy film	Cathodic electrophoretic deposition	0.0469 (2 mV s^{-1})	0.5 M Na_2SO_4	[63]
RPC-0.1	In-situ polymerization and chemical reduction	0.52 (2 mA cm^{-2})	1 M NaCl	This work
RPC-0.5	In-situ polymerization and chemical reduction	0.75 (2 mA cm^{-2})	1 M NaCl	This work
RPC-2.5	In-situ polymerization and chemical reduction	1.20 (2 mA cm^{-2})	1 M NaCl	This work

[a] Solid-stated electrolyte

Table S3. Comparison of areal capacitances and volumetric capacitances of RPC-SSC with other recently reported solid-state supercapacitors.

Electrode	Type	Areal capacitance (F cm^{-2})	Volumetric capacitance (F cm^{-3})	Electrolyte	Ref.
PPy/paper	SSC	0.42 (1 mA cm^{-2})	11 (26.3 mA cm^{-3})	H_3PO_4/PVA	[15]
VO_x//VN	ASC	–	1.35 (0.5 mA cm^{-2})	LiCl/PVA	[68]
TiO_2@MnO_2//TiO_2@C	ASC	–	0.71 (10 mV s^{-1})	LiCl/PVA	[69]
TiN nanowire arrays	SSC	–	0.33 (2.5 mA cm^{-3})	KOH/PVA	[70]
Graphene/PPy composite fibers	SSC	0.115 (0.2 mA cm^{-2})	–	H_2SO_4/PVA	[62]
Graphene–Cellulose Paper	SSC	0.046 (2 mV s^{-1})	–	H_2SO_4/PVA	[7]
RPC-2.5	SSC	0.51 (0.1 mA cm^{-2})	8.5 (1.7 mA cm^{-3})	H_3PO_4/PVA	This work

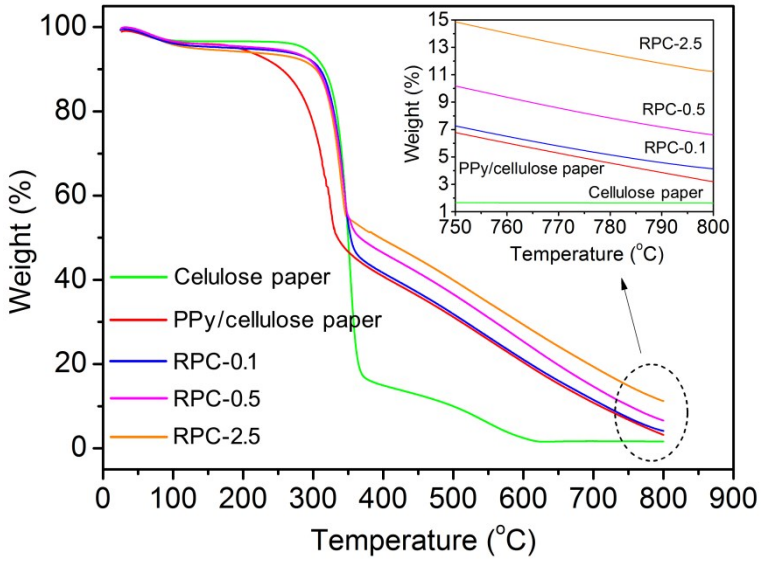

Figure S1. TG curves of the cellulose paper, PPy/cellulose paper, RPC-0.1, RPC-0.5, and RPC-2.5, respectively.

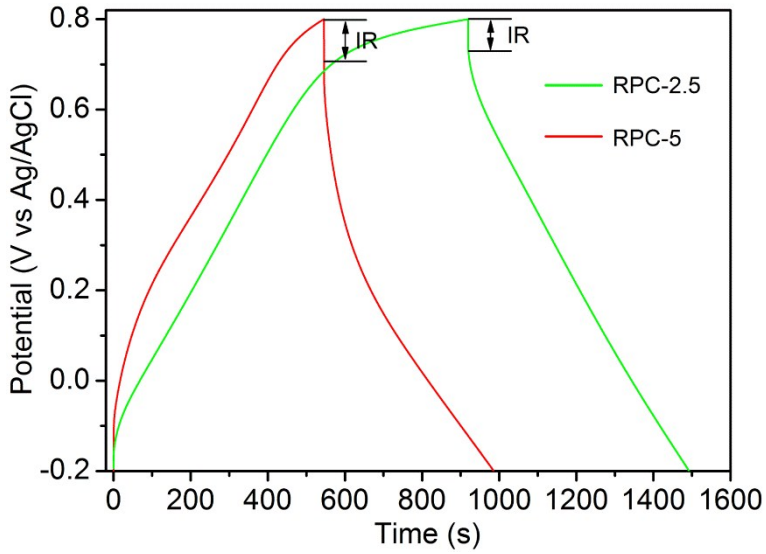

Figure S2. GCD curves of RPC-2.5 and RPC-5 at the current density of 2 mA cm^{-2}.

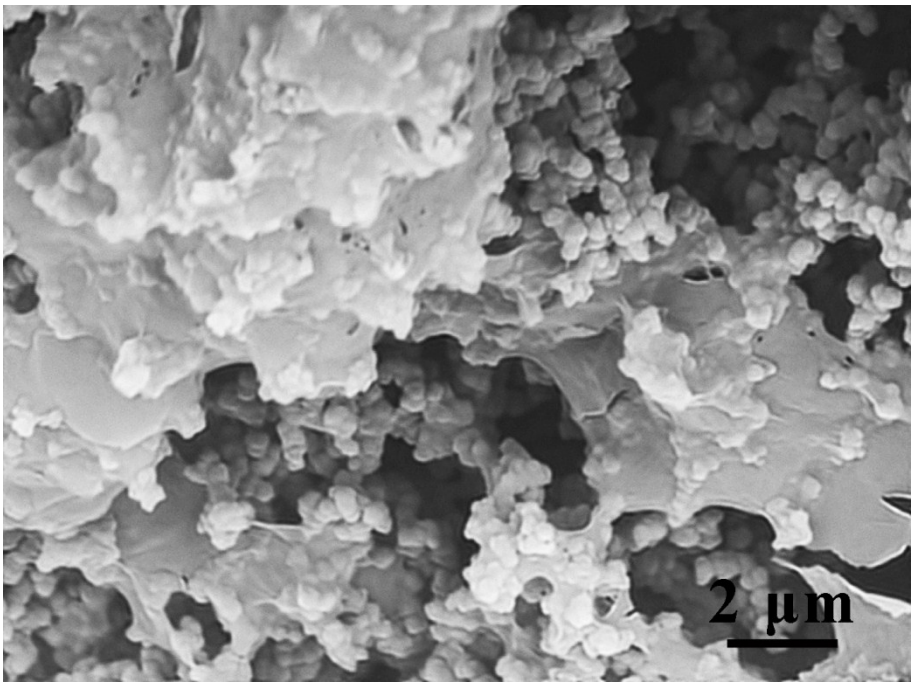

Figure S3. SEM image of RPC-5.

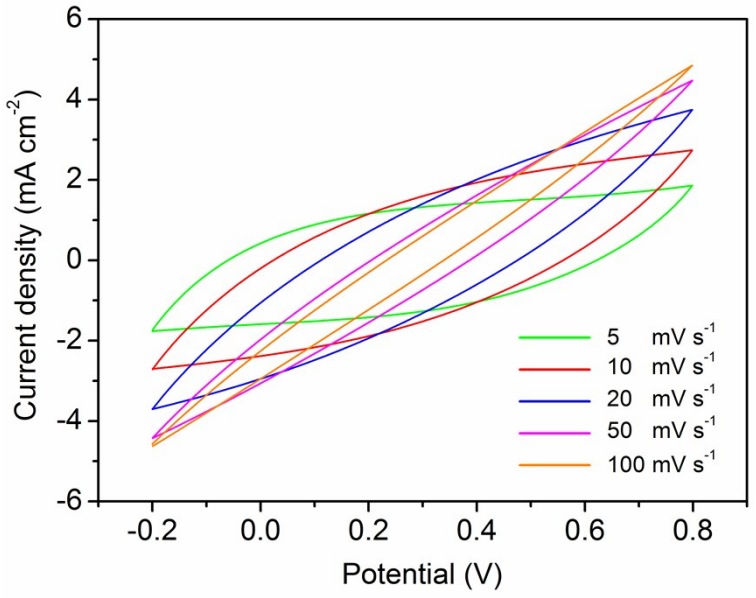

Figure S4. CV curves of RPC-SSC at different scan rates of 5, 10, 20, 50, and 100 mV s^{-1}, respectively.