

Supplementary Material

A monolithic integrated ultra-flexible all-solid-state supercapacitor based on polyaniline conducting polymer

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The specific areal capacitance of single electrode (C_s), energy density (E) and power density (P) could be extracted from the CV and GCD curves by the following equations:

$$C_s = \frac{\oint I dU}{A v \Delta U} \quad S1$$

where v is the CV scan rate, ΔU (~ 1.2 V) is the potential window and A is the electrode area.

$$C_s = \frac{2It}{A \Delta U} \quad S2$$

where I is the constant discharge current, t is the discharge time, ΔU (~ 1.2 V) is the potential window and A is the electrode area.

$$E = \frac{C_s \Delta U^2}{4h} \quad S3$$

where C_s is the specific areal capacitance of single electrode extracted from equation S2, ΔU (~ 1.2 V) is the potential window and h is the whole thickness of the device including the current collectors, electrodes, electrolyte and support.

$$P = \frac{E}{t} \quad S4$$

Table S1 Comparison of several typical membranes used as the SC supports

	Cost	Weight	Porosity	Flexibility
Polycarbonate (PC)	✗	✓	✗	✓
Polyethylene terephthalate (PET)	✓	✓	✗	✓
Carbon nanotube (CNT) or graphene	✗	✓	✓	✓
Polydimethylsiloxane (PDMS)	✓	✓	✗	✓
Polyimide (PI)	✓	✓	✗	✓
Nylon	✓	✓	✓	✓

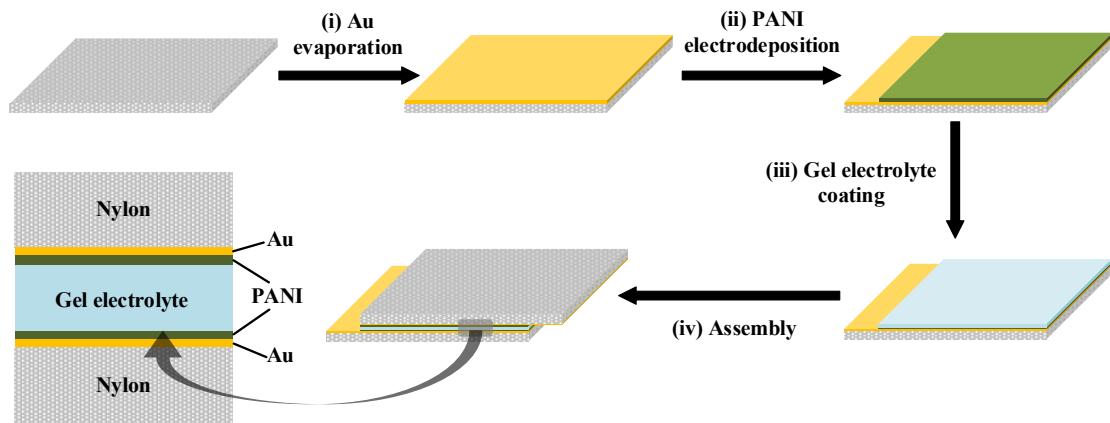


Fig. S1. Fabrication processes of the reference SC.

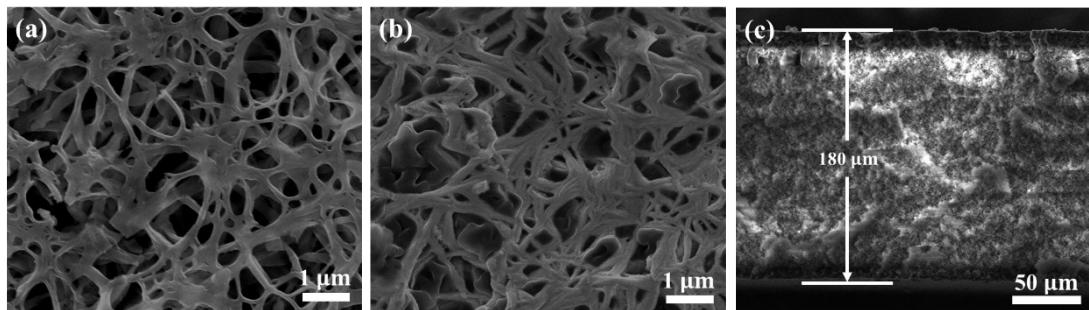


Fig. S2. Plan-view SEM images of the nylon membrane (a) before and (b) after Au deposition; (c) cross-sectional SEM image of the fresh nylon membrane.

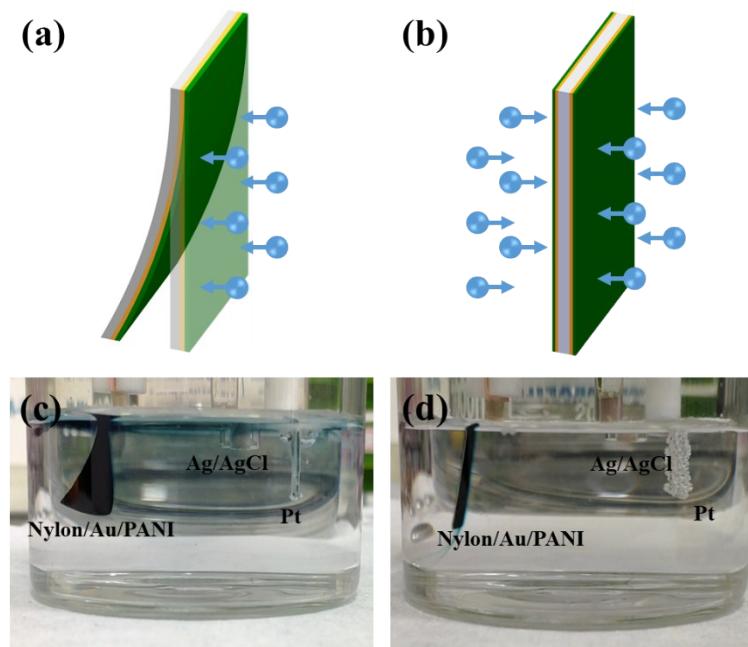


Fig. S3. Schematic diagrams of the PANI electrodeposition on (a) only one side and (b) both sides of the nylon membrane. Photographs of the PANI electrodeposition on (c) only one side and (d) both sides of the nylon membrane.

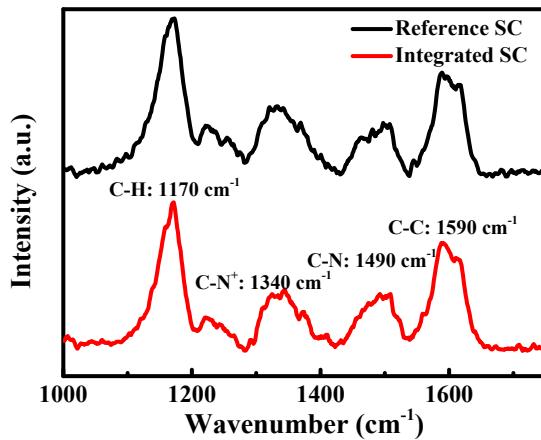


Fig. S4. Raman spectra of the PANI films for the integrated and reference SCs.

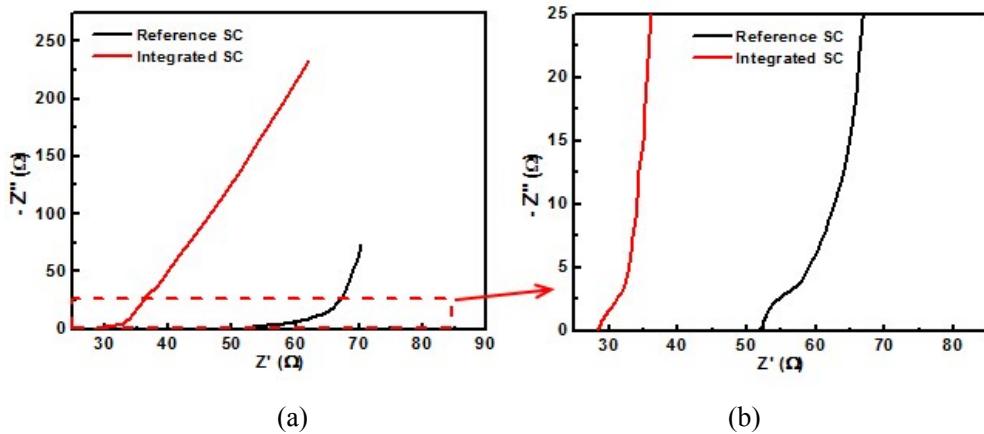
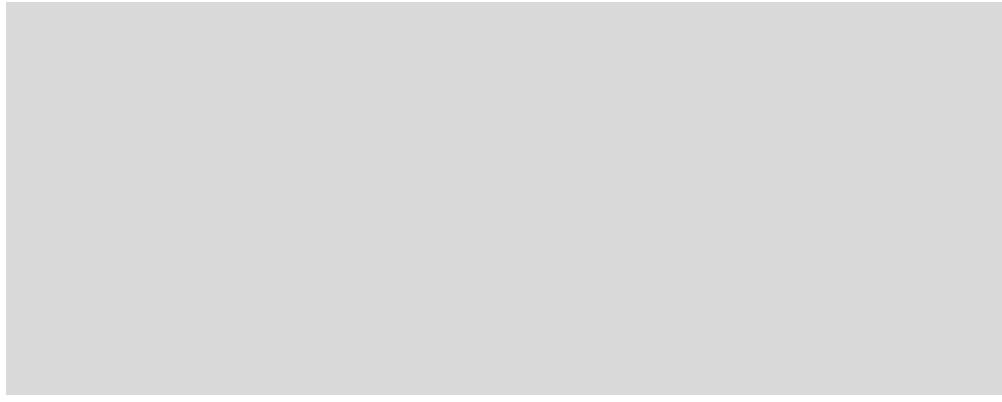


Fig. S5. (a) Nyquist plots of the integrated and reference SCs before cycling and (b) the enlarged Nyquist plots in the rectangular area of Fig. S5(a). As seen in the figure, the equivalent series resistance (ESR) can be extracted from the intersection between the EIS curve and x axis (at $y=0$). The charge transfer resistance (R_{ct}) corresponds to the diameter of the semicircle of the EIS curve at high-to-medium frequencies, and the integrated SC shows much smaller diameter than the reference one, suggesting its much lower R_{ct} .



(a)

(b)

Fig. S6. (a) Nyquist plots of the SCs after cycling and (b) the enlarged Nyquist plots in the rectangular area of Fig. S6(a). Compared with the EIS curves before cycling (as seen in Fig. S5), the EIS curves after cycling degrade for both the integrated and reference SCs. However, the reference SC exhibits much more severe degradation than the integrated one and the ESR before and after cycling is $52.1\ \Omega$ and $62.0\ \Omega$ for the reference SC and $28.6\ \Omega$ and $31.2\ \Omega$ for the integrated SC, corresponding to 19.0% and 9.1% degradation respectively.

Table S2 Performance comparison between the present and previous reported works. Note that the item of capacitance (mF cm^{-2}) represents the specific areal capacitance of single electrode. Regarding the items of power density and energy density, the whole SC volume is used for calculating the energy and power densities in this work and Ref [16-18], while only the electrode volume is used for calculation in Ref [2, 14] and the whole volume except the support is used for calculation in Ref [15].

Composition	Potential range (V)	Capacitance (mF cm^{-2})	Power density	Energy density	Discharge rate (mA cm^{-2})	Capacitance retention
EG-PANI [1]	0~1	80.4	4.14 mW cm^{-2}	$0.003 \text{ mWh cm}^{-2}$	1	90.24% (1000)
PANI [2]	-0.6~0.6	131.3	140 mW cm^{-3}	21.8 mWh cm^{-3}	1	95.2% (2000)
PAN/G/MWCNTs/PPS [3]	0~2	300	4.44 mW cm^{-2}	$0.123 \text{ mWh cm}^{-2}$	4	98% (2000)
PANI/MLG [4]	0~0.7	60	0.15 mW cm^{-2}	0.01 mWh cm^{-2}	0.85	100% (1000)
NG/PANI [5]	0~1	584.7	9.999 mW cm^{-2}	$0.051 \text{ mWh cm}^{-2}$	20	108% (2000)
HGC-PANI [6]	0~1	~450	600 mW cm^{-3}	9.4 mWh cm^{-3}	10	90% (3000)
CNF/CNTs/PANI [7]	0~0.8	67.31	1.92 mW cm^{-2}	$0.006 \text{ mWh cm}^{-2}$	4	90% (5000)
PANI [8]	-0.2~0.8	15	$\sim 0.3 \text{ mW cm}^{-2}$	$0.001 \text{ mWh cm}^{-2}$	0.3	90% (1000)
$\text{Ni}_3\text{V}_2\text{O}_8 @ \text{PANI}$ [9]	0~1.6	52.8	7.06 mW cm^{-2}	$0.016 \text{ mWh cm}^{-2}$	3	88% (20000)
CNFs/CNTs/PANI [10]	-0.2~0.8	626	2.5 mW cm^{-2}	0.05 mWh cm^{-2}	5	71% (10000)
CNT/PANI [11]	0~0.7	184.6	1.18 mW cm^{-2}	$0.008 \text{ mWh cm}^{-2}$	5	95% (500)
PANI/CNT/EVA [12]	0~0.8	192.3	1200 mW cm^{-2}	8.35 mWh cm^{-2}	5	66.4% (3000)
PHE-PANI [13]	0~0.8	131	0.4 mW cm^{-2}	$0.012 \text{ mWh cm}^{-2}$	1	83.7% (5000)
(RGO/CNTs)@PANI [14]	0~0.8	36.7 F cm^{-3}	200 mW cm^{-3}	0.8 mWh cm^{-3}	2 A cm^{-3}	80.6% (2000)
ACM/MWCNT/PANI [15]	0~2.5	2.2 F cm^{-3}	500 mW cm^{-3}	1.13 mWh cm^{-3}	30	80.3% (5000)
a- $\text{Fe}_2\text{O}_3/\text{PANI}$ [16]	0~1.8	236.8	67 mW cm^{-3}	0.31 mWh cm^{-3}	10 A g^{-1}	80.3% (5000)
PANI/MWCNT/PDMS [17]	0~0.8	2.1 F cm^{-3}	185 mW cm^{-3}	0.09 mWh cm^{-3}	6	95% (500)
PANI [18]	0~0.7	718	1.47 mW cm^{-3}	$0.051 \text{ mWh cm}^{-3}$	0.5	96% (13000)
Reference SC	-0.6~0.6 V	308.5	17.4 mW cm^{-3} (1.2 mW cm^{-2})	0.2 mWh cm^{-3} (0.02 mWh cm^{-2})	2	83.7% (6500)
Integrated SC	-0.6~0.6 V	341.7	73.9 mW cm^{-3}	1.7 mWh cm^{-3}	2	92.8% (6500)

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