

## Supporting Information

### Preparation of Polyaniline/Polyurethane Hydrogels for High-performance All-in-one Supercapacitor with Remarkable Pressure-Resistance

*Pu Hou<sup>a,b</sup>, Zishuo Li<sup>a,b</sup>, Jiefei Liu<sup>a,b</sup>, Haoxiang Zhang<sup>a,b</sup>, Dawei Zhang<sup>a,b</sup>, Jidong*

*Dong<sup>a,b</sup>, Jiyou Gu<sup>a,b\*</sup>, Pengfei Huo<sup>a,b\*</sup>*

<sup>a</sup> State Key Laboratory of Woody Oil Resources Utilization, Northeast Forestry University, Harbin, 150040, China

<sup>b</sup> College of Materials Science and Engineering, Northeast Forestry University, Harbin 150040, China

**\*Email: [huopengfei@nefu.edu.cn](mailto:huopengfei@nefu.edu.cn)**

**\*Email: [dldgujy@nefu.edu.cn](mailto:dldgujy@nefu.edu.cn)**

#### Equations

$$i = av^b \tag{S1}$$

where  $I$  and  $v$  are the peak current (mA) and scan rates (mV s<sup>-2</sup>), respectively. Take the logarithm of the formula:  $\log(i) = \log(a) + b\log(v)$ , and the slope obtained by fitting is  $b$  value.

$$\sigma = \frac{L}{R \times A} \tag{S2}$$

where  $L$  (cm),  $R$  ( $\Omega$ ) and  $A$  (cm<sup>2</sup>) are the thickness of hydrogel, bulk resistance and contact area, respectively.

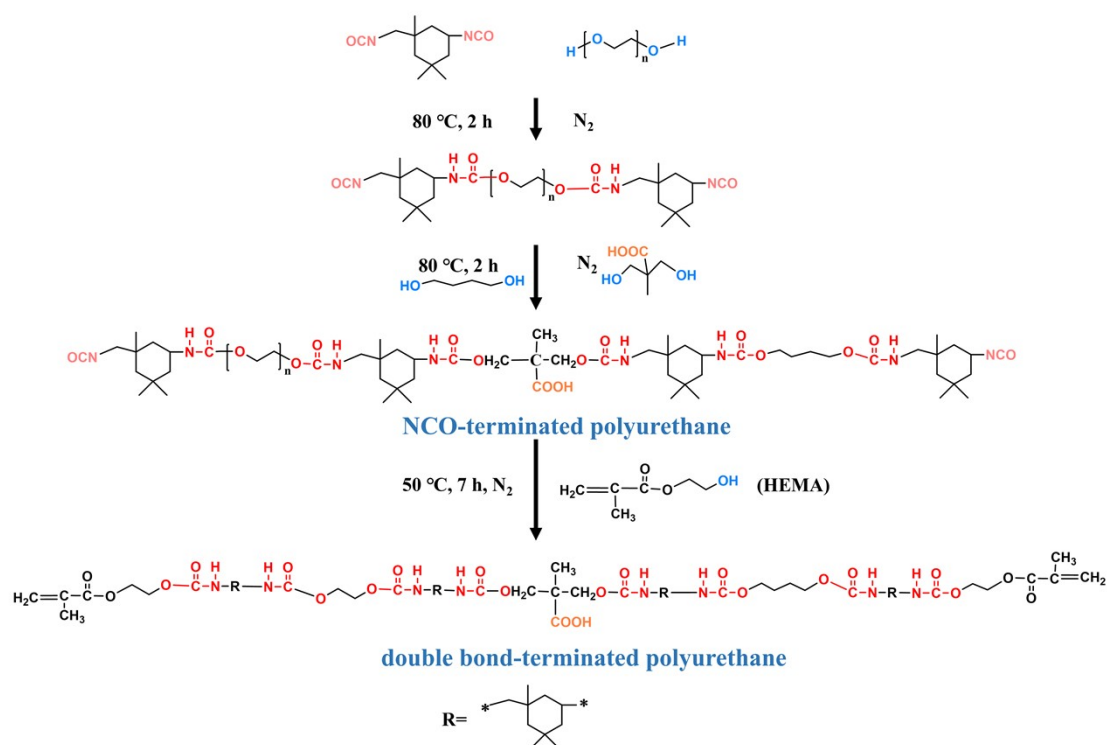


Figure S1. Synthesis of double bond-terminated polyurethane (HPU)

Table S1. The addition amounts of materials of PAM-PU-PAMPS/CA terpolymers

Sample designation	Composition of PAM-PU-PAMPS/CA terpolymers					
	AMPS (g)	HPU (g)	CA (10 wt%)	AIBN (1.0 wt%)	AM (g)	HPU/AMPS/AM (mass ratio)
3A-PU-S/C	1.50	0.50	0.05	0.035	1.5	1:3:3
4A-PU-S/C	1.50	0.50	0.05	0.040	2.0	1:3:4
5A-PU-S/C	1.50	0.50	0.05	0.045	2.5	1:3:5
6A-PU-S/C	1.50	0.50	0.05	0.051	3.0	1:3:6

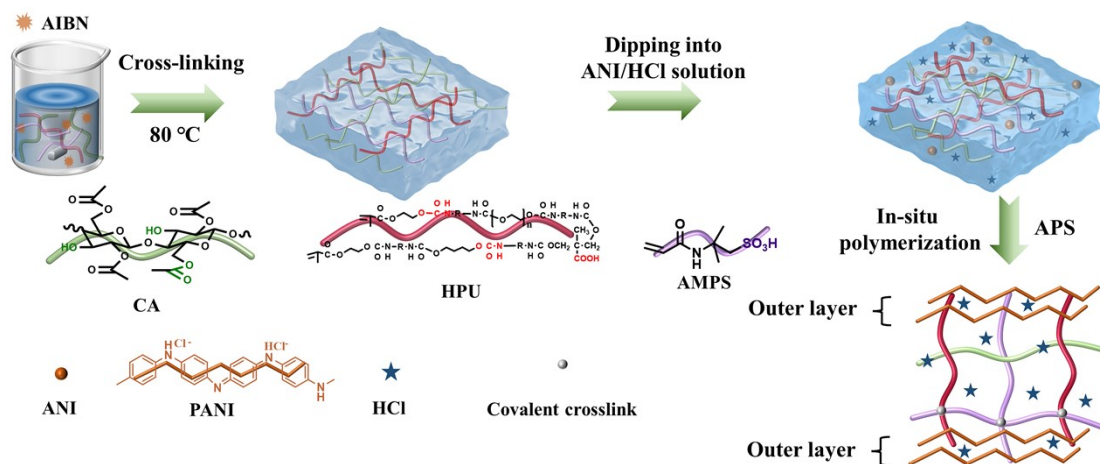


Figure S2. Synthetic pathway of PANI/PU-S/C.

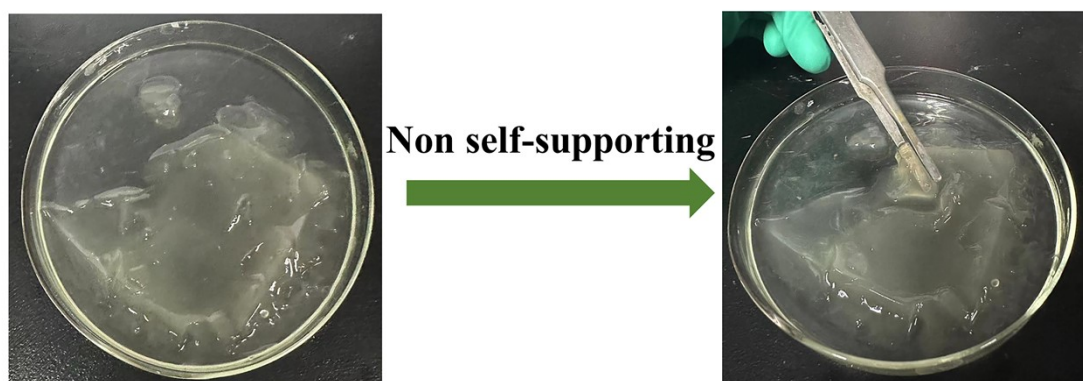


Figure S3. Photograph of over swollen PU-S/C hydrogel.

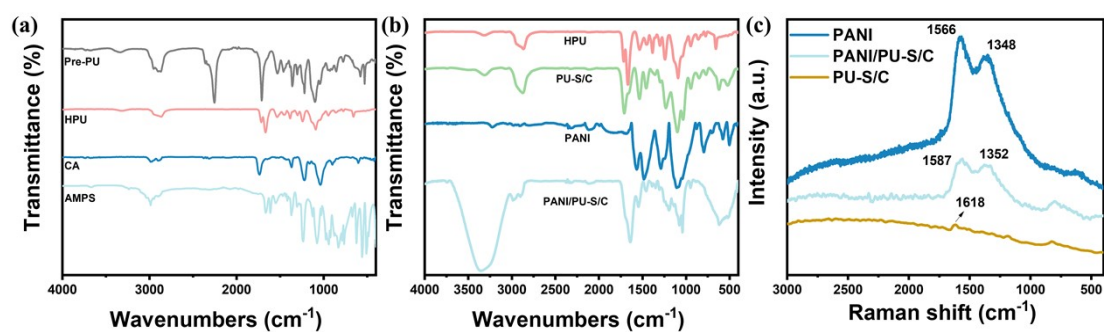


Figure S4. FTIR spectra of (a) Pre-PU, HPU, CA and AMPS monomer; (b) HPU, PU-S/C, PANI/PU-S/C and pure PANI; (c) Raman spectra of pure PANI, A-PU-S/C composite and PANI/PU-S/C.

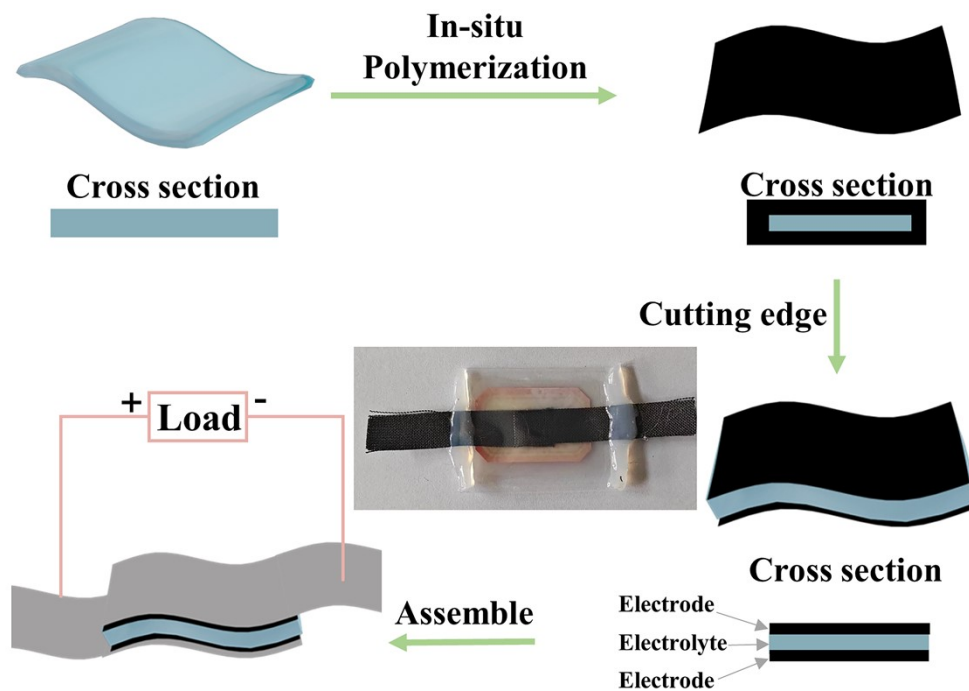


Figure S5. The assembly of PANI/PU-S/C based all-in-one SCs.

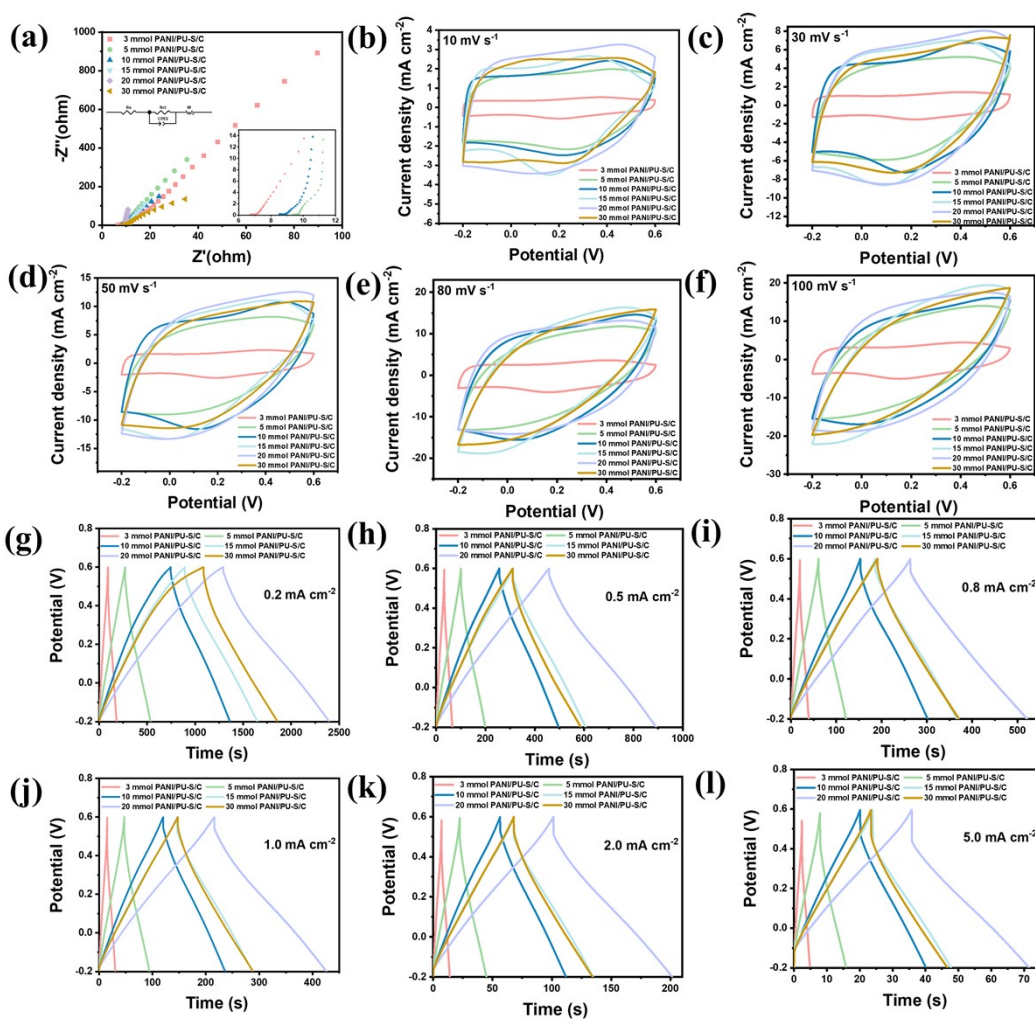


Figure S6. Electrochemical characterization of the PANI/PU-S/C based all-in-one SCs with various aniline concentrations. (a) Nyquist plots of PANI/PU-S/C based all-in-one SCs; (b-f) CV curves of the PANI/PU-S/C based all-in-one SCs at 10-100 mV s<sup>-1</sup>; (g-l) GCD curves of the PANI/PU-S/C based all-in-one SCs at 0.2-5.0 mA cm<sup>-2</sup>.

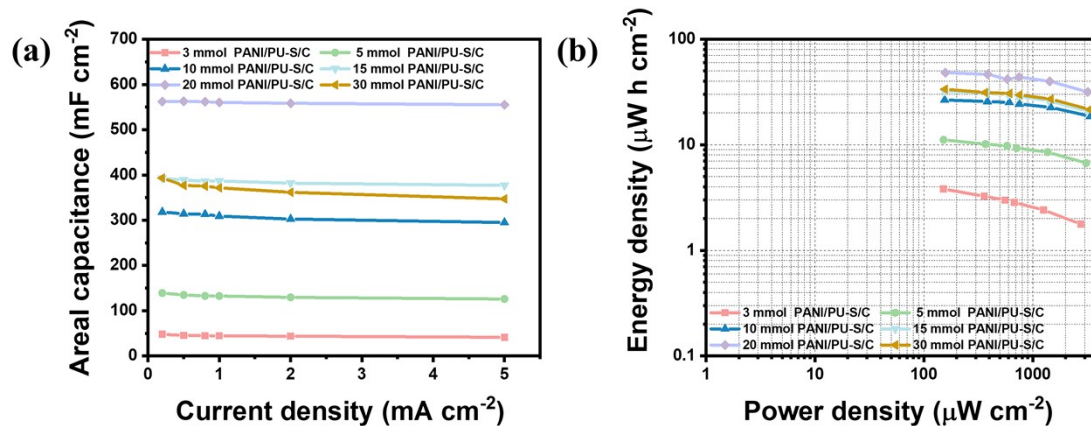


Figure S7. The areal capacitances (a) and the Ragone plots (b) of the PANI/PU-S/C based all-in-one SCs with different content of aniline at different current densities.

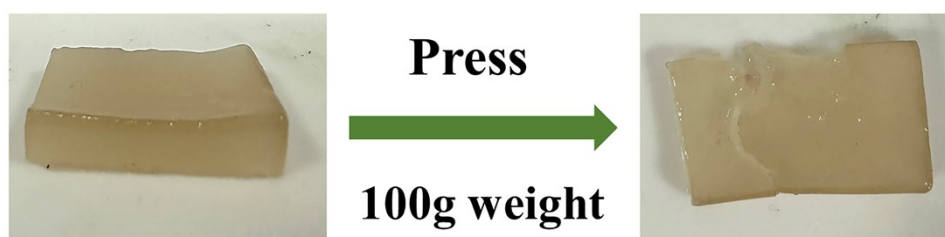


Figure S8. The photographs of PU-S/CA hydrogel before and after being pressed

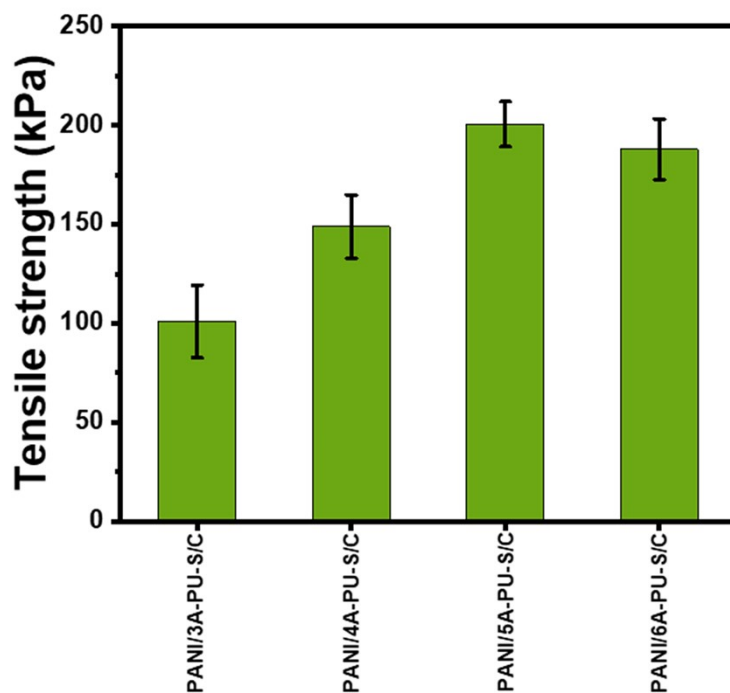


Figure S9. The tensile strength bar chart of PANI/A-PU-S/C-based D-CPH

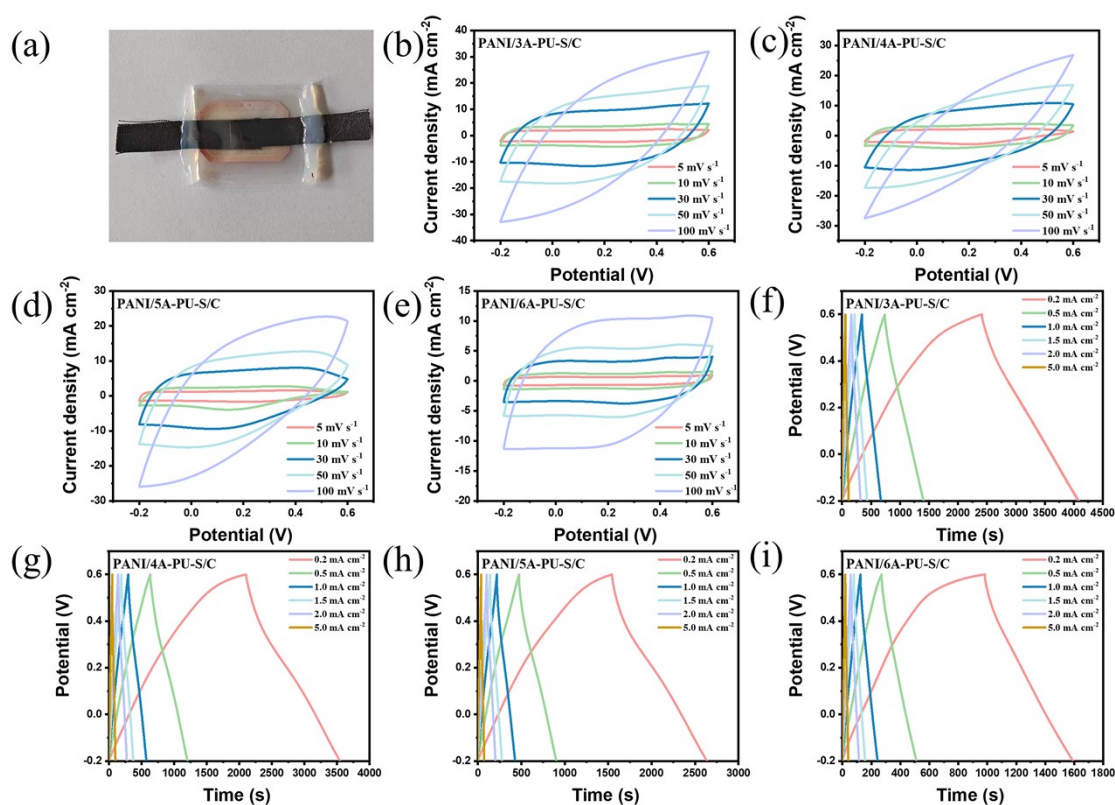


Figure S10. (a) The photograph of assembled PANI/A-PU-S/C based all-in-one SC; (b-e) CV curves of the PANI/A-PU-S/C based all-in-one SCs with different AM contents at scan rates of 5-100  $\text{mV s}^{-1}$ ; (f-i) GCD curves of the PANI/A-PU-S/C based all-in-one SCs with different AM contents at current density of 0.2-5.0  $\text{mA cm}^{-2}$ .

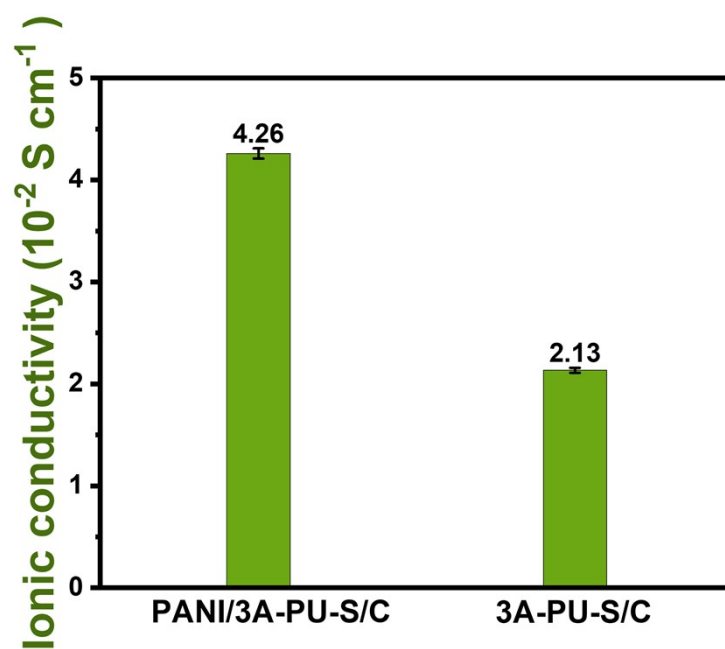


Figure S11. The ionic conductivity of the pure 3A-PU-S/C-based hydrogel and PANI/3A-PU-S/C based CPH.

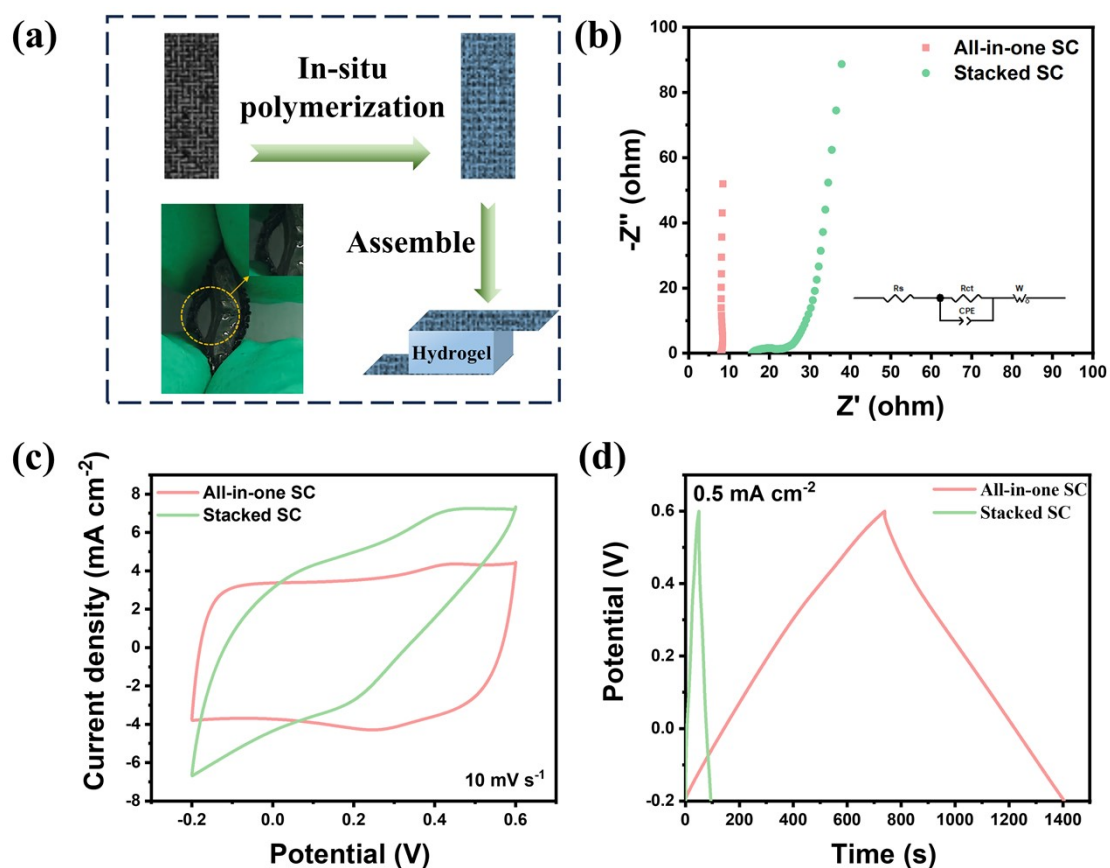


Figure S12. (a) The assembly process of stacked structure supercapacitor using PANI/carbon cloth as electrodes (The illustration is the bent state stacked SC); (b) EIS plots of the all-in-one SC and the stacked SC; (c) CV curves of the all-in-one SC and the stacked SC at scan rate of  $10 \text{ mV s}^{-1}$ ; (d) GCD curves of the all-in-one SC and the stacked SC at current density of  $0.5 \text{ mA cm}^{-2}$ .

Table S2. Comparison of the electrochemical performance of reported advanced CPHs-based supercapacitors with this work.

Sample	$C_a$ (mF cm <sup>-2</sup> )	$E_a$ (μW h cm <sup>-2</sup> )	$P_a$ (μW cm <sup>-2</sup> )	Ref.
LS-PEDOT/PAAM/Fe <sup>3+</sup>	672.5 (1.0 mA cm <sup>-2</sup> )	60	800	[3]
Poly (EDOT-co-(Th-g-PAA)-co-ThBA)-PVA	335.40 (10 mA cm <sup>-2</sup> )	19.8	-	[10]
PHEA-g-PPy	430 (0.2 mA cm <sup>-2</sup> )	38.2 29	91.4 1371	[20]
PPy/PAM/LiCl	229.8 (0.8 mA cm <sup>-2</sup> )	20.4 16	634.7 1600	[21]
PAM/SA/PLum/Pom	134.6 (2 mA cm <sup>-2</sup> )	33.65	2000	[22]
PVA/PHEA/PANI	98 (0.2 mA cm <sup>-2</sup> )	8.48	78.52	[28]
PAM-PVA/HACC	250.75 (0.5 mA cm <sup>-2</sup> )	23.9	250	[30]
P(AM-co-AA)/Trehalose/LiCl	148.2 (0.5 mA cm <sup>-2</sup> )	20.04	260	[37]
PPy/PVP/PAM/LiCl	308 (1.0 mA cm <sup>-2</sup> )	21.1	701.4	[38]
AQS-PEDOT/PAA/H <sub>2</sub> SO <sub>4</sub> +KCl	466.5(1.0 mA cm <sup>-2</sup> )	41.47	400	[39]
PVA/PA-PANI	356.5 (1.0 mA cm <sup>-2</sup> )	31.7	800	[40]
PAD/H <sub>2</sub> SO <sub>4</sub> -PANI	430 (0.5 mA cm <sup>-2</sup> )	22	150	[41]
P(ANI-co-Py)/ PVA	317 (1 mA cm <sup>-2</sup> )	44	250	[42]
Gel/PAAM/PPy	363.7 (0.5 mA cm <sup>-2</sup> )	32.33	400	[43]
PPy /PVA/KCl	224 (0.8 mA cm <sup>-2</sup> )	20 13	600 2300	[44]
PANI/3A-PU-S/C	837.53 (5 mA cm <sup>-2</sup> )	53.13	3379	This work

Table S3. Comparison of the performance data of previously reported CPH-based all-in-one SCs with this work.

<b>Conductive polymer matrix</b>	<b>Compressive stress (kPa)</b>	<b>Compressive strain (%)</b>	<b>Tensile strength (kPa)</b>	<b>Energy density (<math>\mu\text{W h cm}^{-2}</math>)</b>	<b>Power density (<math>\mu\text{W cm}^{-2}</math>)</b>	<b>Ref.</b>
PPy/PVA/AA	90	60	173.9	14.9	300	[48]
PPy/CNT/PAA	5000	60	160	0.048	-	[49]
PANI/PAM	25	50	-	16.4	85.4	[50]
PANI/PAA/O MA	2.5 kg weight	-	2200	-	-	[47]
PANI/PVA-PA	-	-	5400	7.58	-	[51]
PANI/PPHP- Na	221	80	601	34.8	-	[52]
PANI/3A-PU- S/C	Car rolling (~200- 300kPa)	-	115.4	53.13	3379	This work