

## Supplementary Materials for

### **In situ fabricated quasi-solid polymer electrolyte incorporated by ionic liquid for flexible supercapacitor**

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## Supplementary Notes

The ionic conductivity of the QSPE was calculated according to Eq. (1)<sup>[1]</sup>:

$$\sigma = \frac{L}{R \times A} \quad (1)$$

Where  $L$  and  $A$  is the thickness and area of the QSPE, respectively, and  $R$  is the bulk resistance from EIS measurement of the symmetrical stainless steel cell.

The relationship between  $\log\sigma$  and  $T^{-1}$  in Fig.2c was linearly fitted and then activation energy ( $E_a$ ) of the QSPE was obtained according to Eq. (2)<sup>[2]</sup>:

$$\sigma(T) = A \exp\left(-\frac{E_a}{kT}\right) \quad (2)$$

Where  $A$  is pre-exponential factor,  $k$  is Boltzmann constant, and  $T$  is the absolute temperature.

The ion diffusion coefficient  $D$ , exchange current density  $i_0$  and time constant  $\tau$  was calculated by Eq. (3), Eq. (4) and Eq. (5), respectively, based on the EIS result of the capacitor<sup>[3-5]</sup>:

$$D = \frac{R^2 T^2}{2A^2 n^4 F^4 C^2 \sigma_0^2} \quad (3)$$

Where  $R$  is the ideal gas constant,  $F$  is the Faraday constant,  $T$  represents the absolute temperature,  $A$  is the electrode area,  $n$  is the number of transferred electrons per molecule during the electrode reaction,  $C$  is the molar concentration of electrolyte,  $\sigma_0$  is the Warburg impedance.

$$i_0 = \frac{RT}{nFAR_{ct}} \quad (4)$$

Where  $R_{ct}$  is the charge transfer resistance, obtained by the diameter of the semicircle in the high-frequency region of the EIS.

$$\tau = \frac{L^2}{D} \quad (5)$$

Where  $L$  is the ion diffusion distance approximating the electrolyte thickness between two electrodes, and  $D$  is the ion diffusion coefficient.

The specific capacitance  $C_s$  of the single electrode was calculated according to Eq. (6), based on charge/discharge test of the capacitor<sup>[6]</sup>:

$$C_s = \frac{I \Delta t}{m \Delta V} \times 4 \quad (6)$$

Where  $m$  is the total mass of two electrodes,  $I$  is the discharge current,  $\Delta t$  is the discharge time, and  $\Delta V$  is discharge voltage interval after removing the IR drop.

The  $C_s$  value can also be obtained according to Eq. (7), based on the CV measurement of the capacitor [7]:

$$C = \frac{\int IdV}{2mv\Delta V} \quad (7)$$

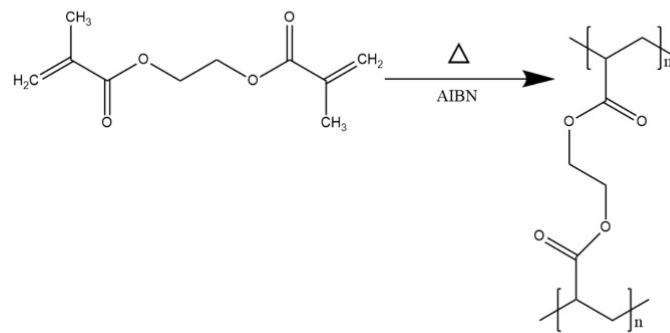
Where the integral part represents the area inside the CV,  $m$  is the total mass of the two electrodes,  $v$  is the scan rate and  $\Delta V$  is the operating voltage window.

The specific energy  $E$  (Wh kg<sup>-1</sup>) and specific power  $P$  (W kg<sup>-1</sup>) of the capacitor was calculated according to Eq. (8) and Eq. (9), respectively [8]:

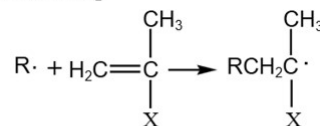
$$E = \frac{1}{8 \times 3.6} C_s \Delta V^2 \quad (8)$$

$$P = \frac{E}{\Delta t} \times 3600 \quad (9)$$

## Supplementary Figures



**Initiation step**



**Propagation step**

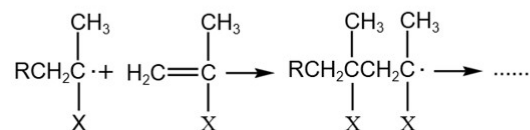


Fig.S1 Free radical polymerization mechanism of PEGDMA induced by AIBN under thermal condition

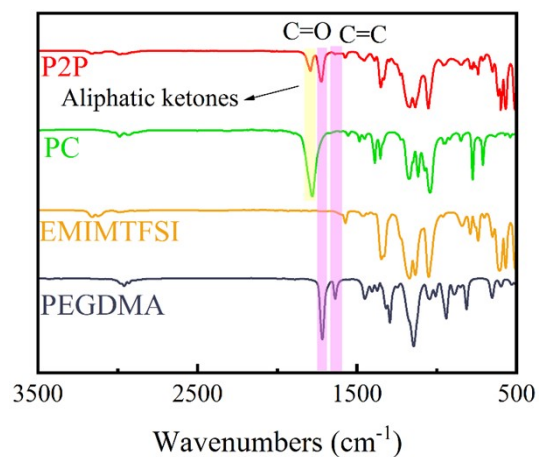


Fig.S2 FTIR spectra of P2P and its corresponding components

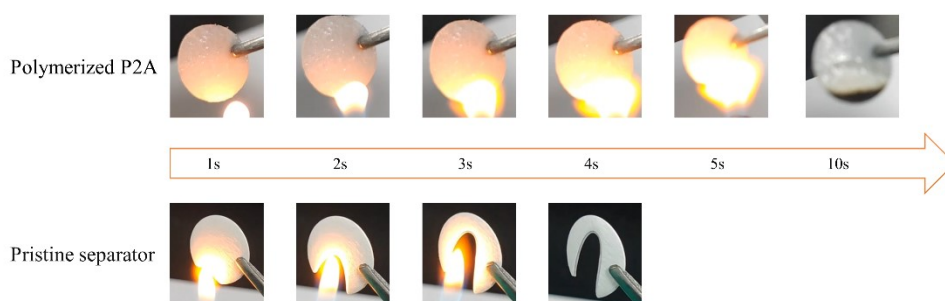


Fig.S3 Pictures of the ignition test on the P2A and cellulose separator

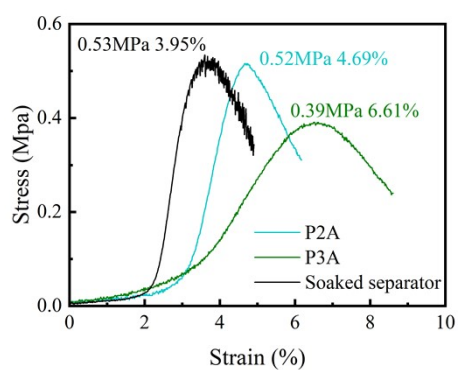


Fig.S4 Stress-strain curves of P2A, P3A and cellulose separator soaked in the IL

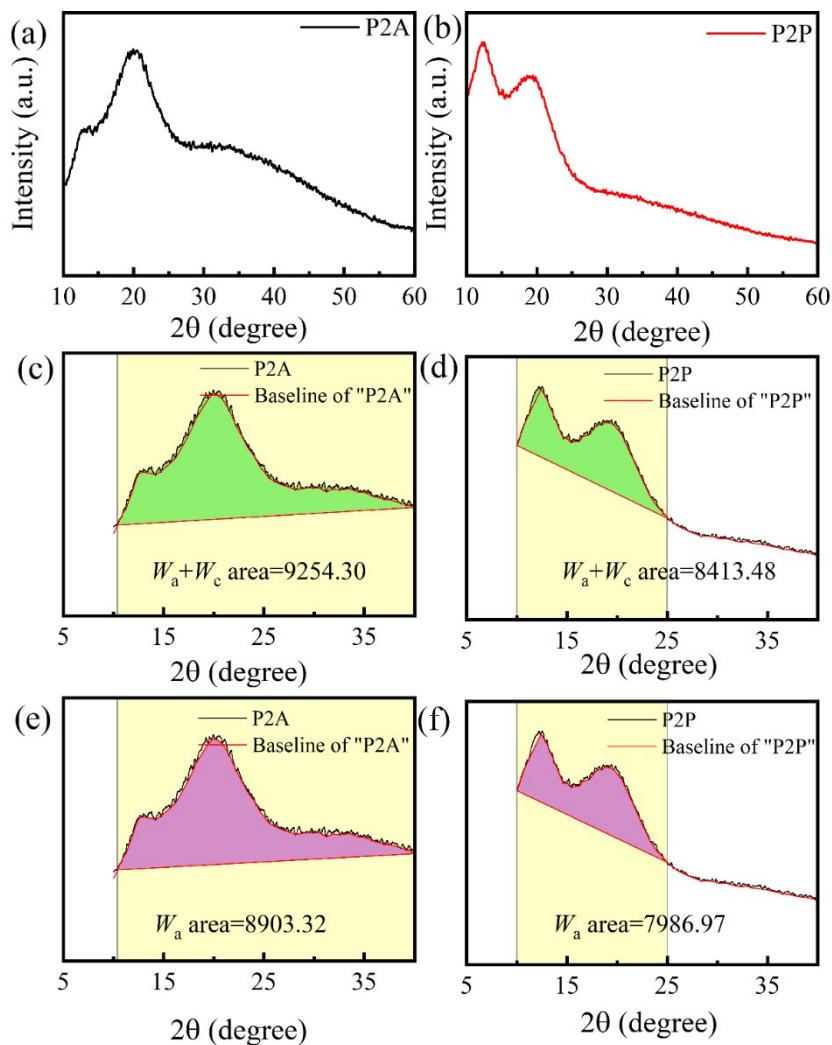


Fig.S5 XRD patterns of (a, c, e) P2A and (b, d, f) P2P, (c, d, e, f).  $W_c$  and  $W_a$  represent the area of the crystalline and amorphous region, respectively. The crystallinity fraction  $X_c$  was calculated as follows [9]:

$$X_c = \frac{W_c}{W_c + W_a} * 100\%$$

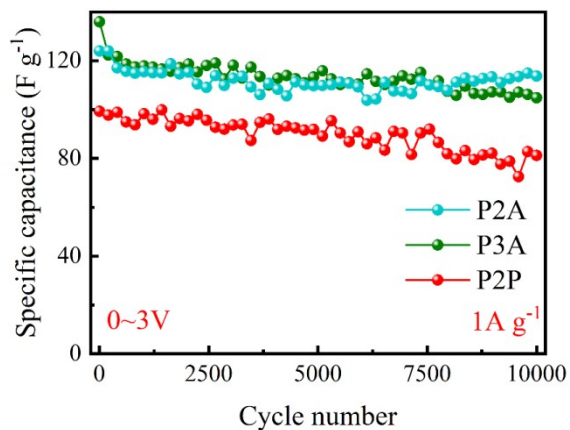


Fig.S6 Comparison of cycle performance of the SCs with three ILQSEs at  $1A g^{-1}$

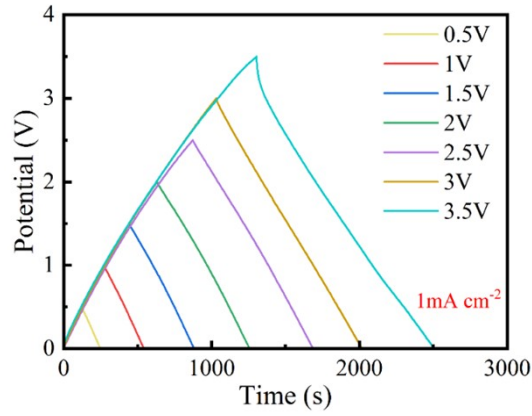


Fig.S7 GCD curves of the SC with P2A at different cut-off voltages

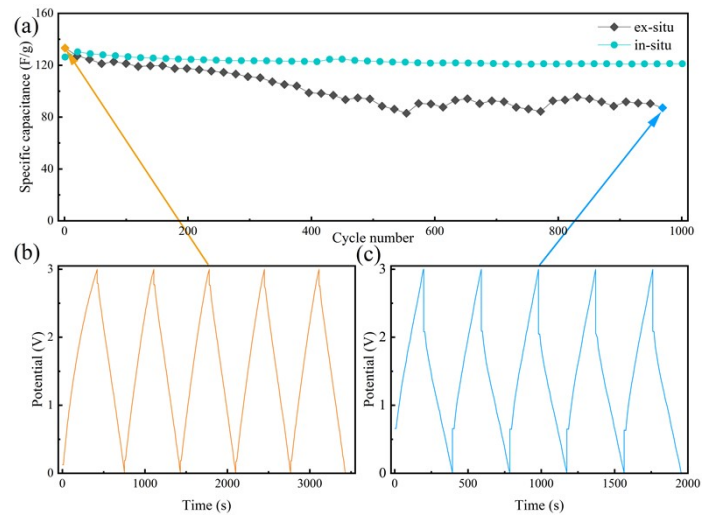


Fig.S8 (a) Cycle performances of the SCs with *in-situ* and *ex-situ* prepared P2A; Charge-discharge curves of the SCs with *ex-situ* prepared P2A at (b) initial cycles and (c) later cycles

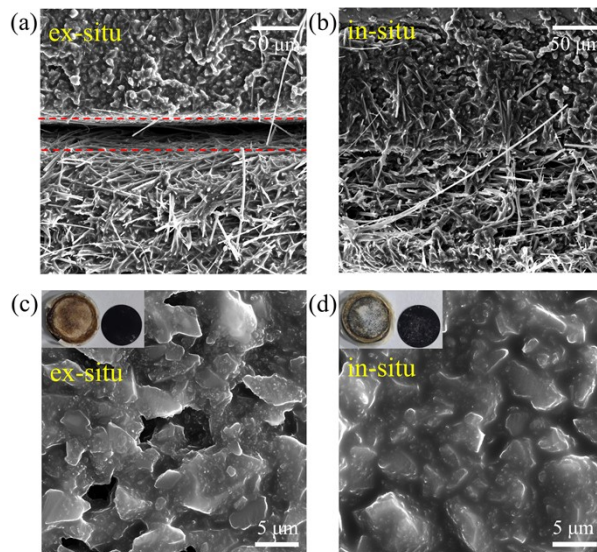


Fig.S9 Cross-section images of the cycled AC electrodes adhered by the (a) *ex-situ* prepared P2A

and (b) *in-situ* prepared P2A; Surface morphologies of the cycled AC electrodes after (c) *ex-situ* prepared P2A and (d) *in-situ* prepared P2A were peeled off from the electrode surface

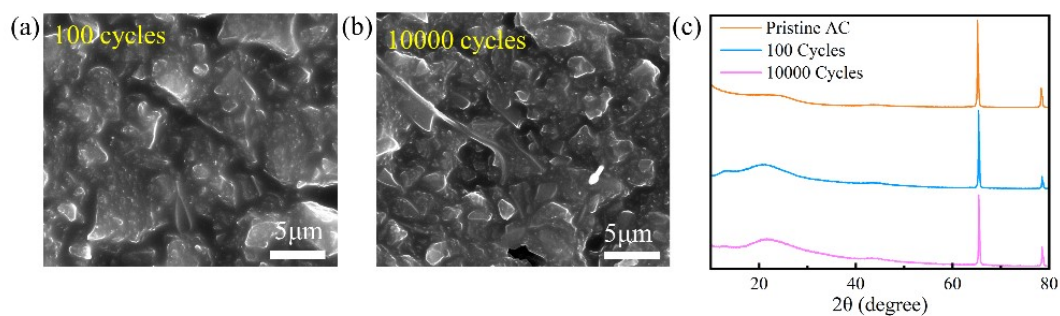


Fig.S10 SEM images of the AC electrode (a) after 100 cycles and (b) after 10000 cycles; (c) Comparison of XRD patterns of the AC electrode during cycling (the *in-situ* prepared P2A were peeled off from the cycled electrode)

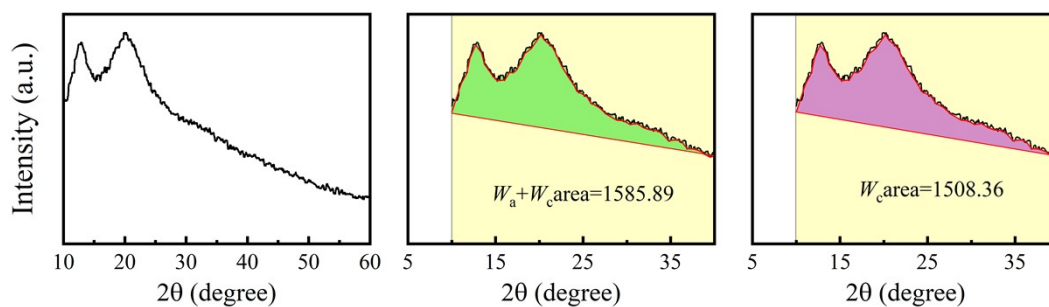


Fig.S11 XRD patterns of the *in-situ* prepared P2A peeled off from the cycled AC electrode

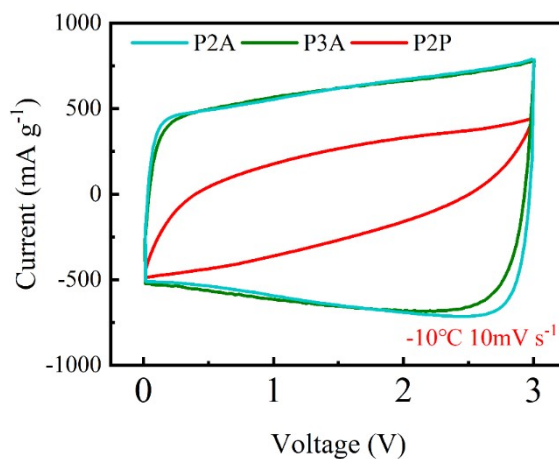


Fig.S12 CV curves of the SCs with three ILQSEs at -10°C

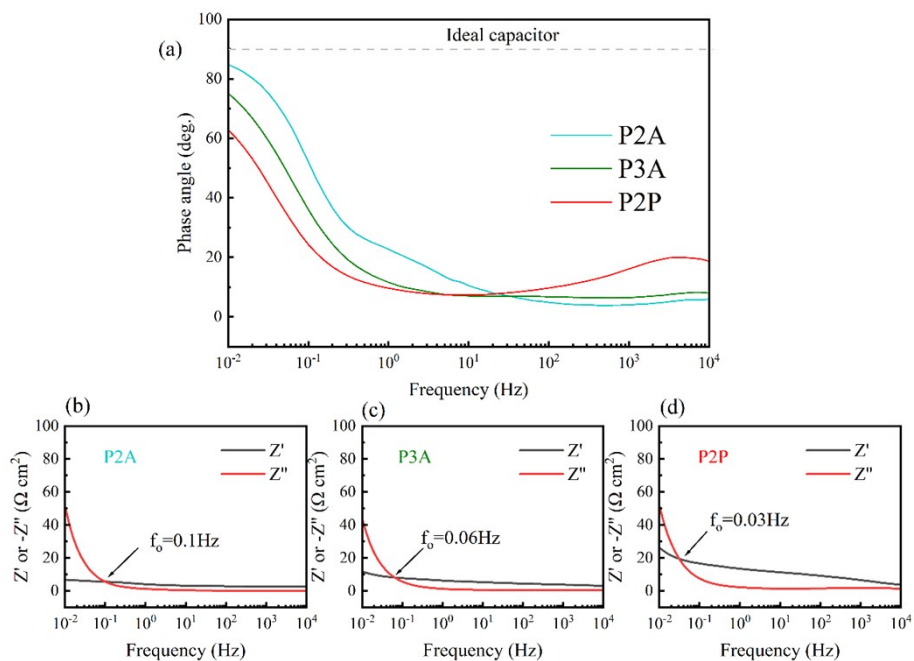


Fig.S13 (a) Phase angle as a function of frequency in the SCs with three ILQSEs; (b) Bode plots ( $Z'$  or  $Z''$  as a function of frequency) of the SCs with (b) P2A, (c) P3A and (d) P2P, respectively

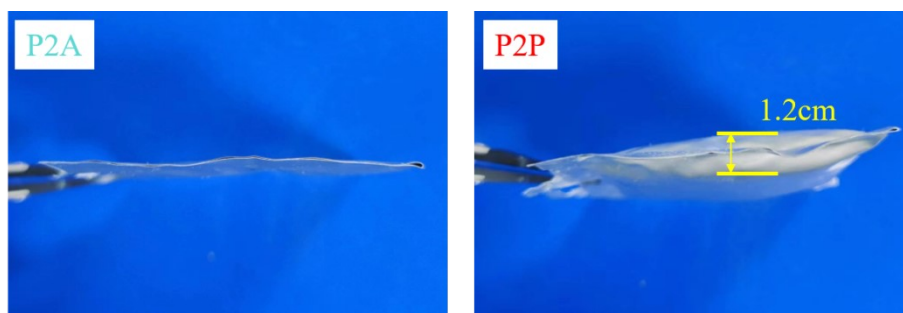


Fig.S14 Cross-section pictures of the flexible SCs after 10000 cycles

## Supplementary Tables

Table S1 Several parameters obtained from the EIS measurements of the SCs

Sample	$R_b$ / $\Omega$	$R_{ct}$ / $\Omega$	$\sigma_0$ / $\Omega$	$D$ / $\text{cm}^2 \text{s}^{-1}$	$i_0$ / $\text{mA cm}^{-2}$	$\tau$ / $\text{s}$	$f_0$ / $\text{Hz}$	$\tau_0$ / $\text{s}$
P2A	2.318	0.386	3.482	1.526	0.043	0.011	0.1	10
P3A	2.488	1.057	4.884	0.776	0.016	0.022	0.06	16.67
P2P	2.497	8.834	5.012	0.319	0.002	0.053	0.03	33.34



## Additional References

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