

## Supporting Information

### **Electrolyte-as-Binder Strategy Using In-Situ-Formed PDOL Gel for Binder-Free Cathodes in Flexible Quasi-Solid-State Li–S Batteries**

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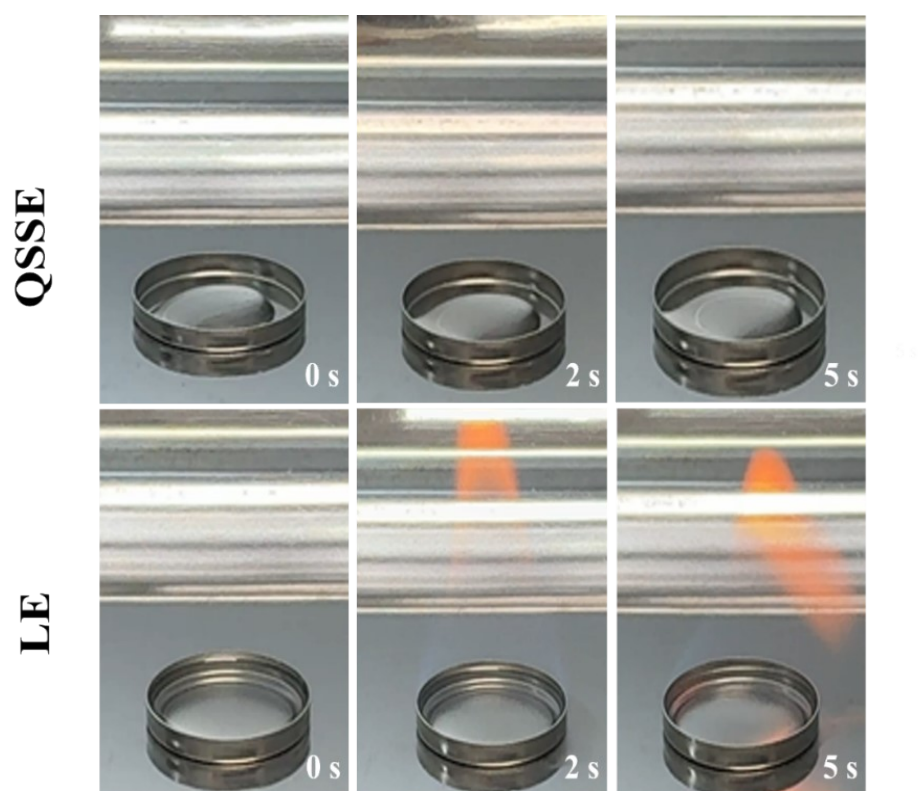
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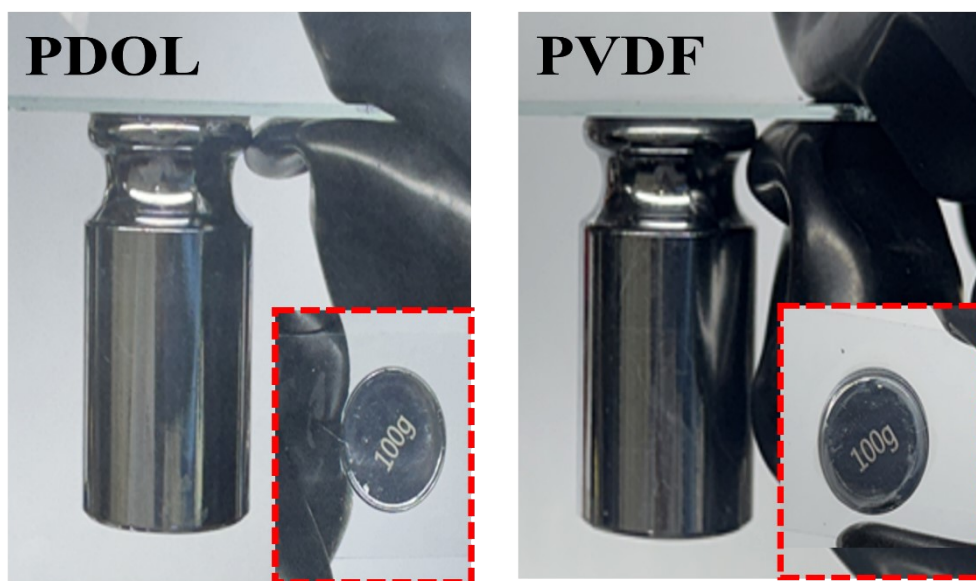
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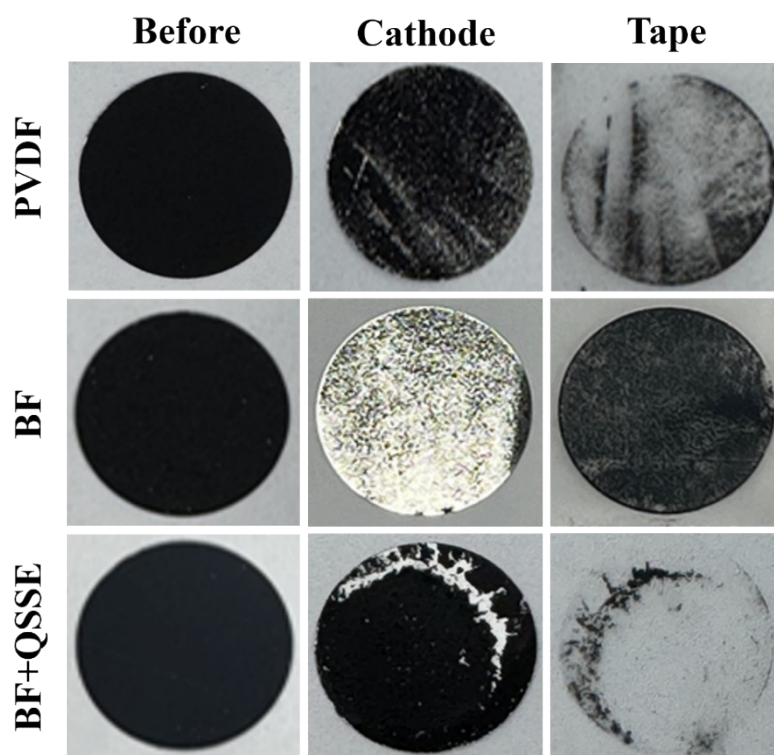
† These authors contributed equally to this work.



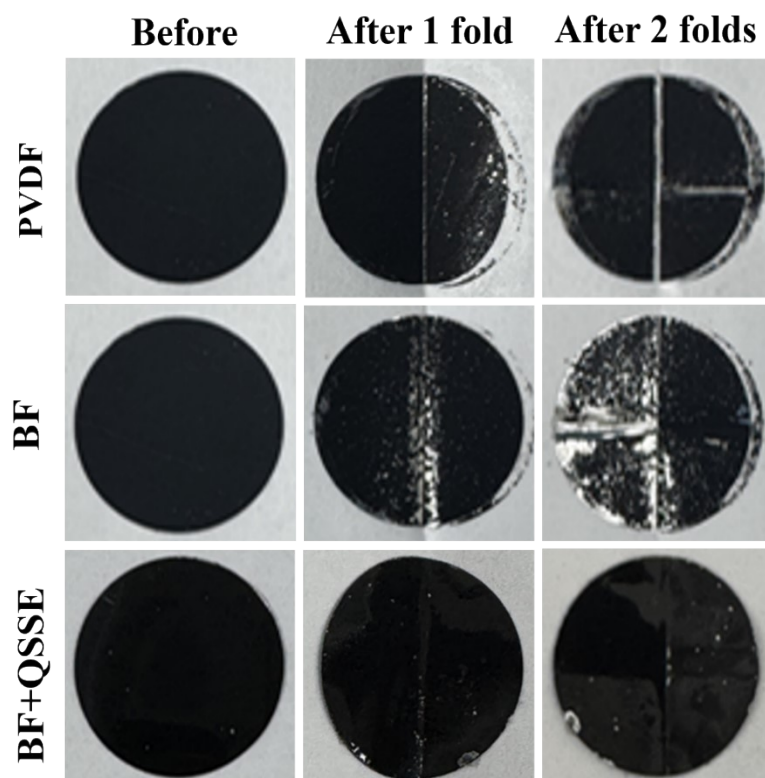
**Figure (S1).** Flammability test of QSSE and LE.



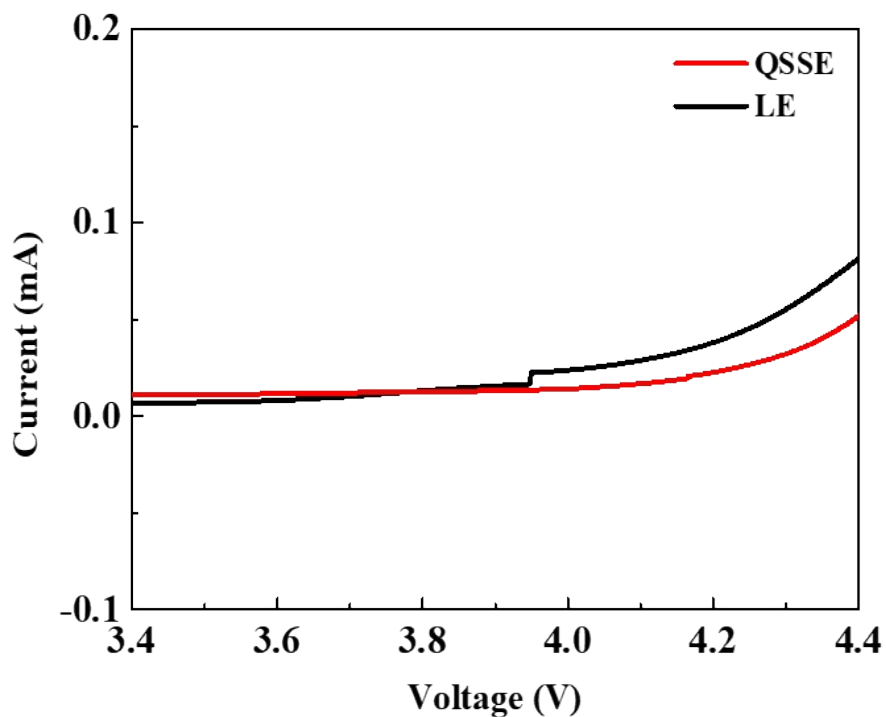
**Figure (S2).** Optical photos of adhesion strength test of PDOL and PVDF.



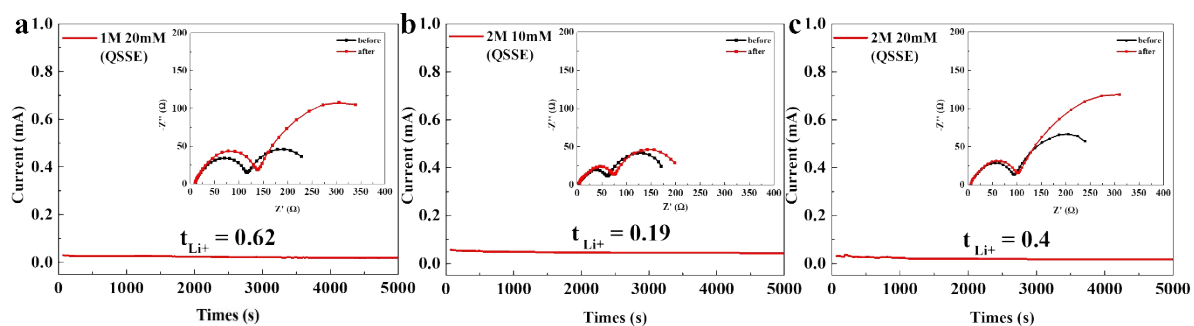
**Figure (S3).** Optical photos of peel tests with cathodes of different cathodes.



**Figure (S4).** Optical photos of folding tests with cathodes of different cathodes.



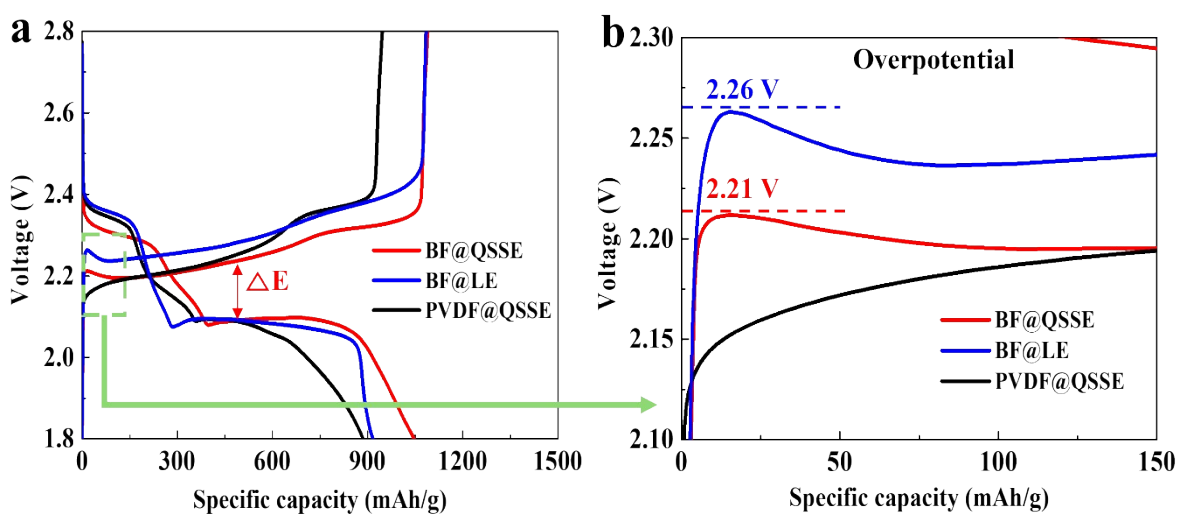
**Figure (S5).** LSV curves of QSSE and LE measured in the voltage range of 3.4–4.4 V.



**Figure (S6).** Steady-state polarization curve of the Li/Li symmetric batteries assembled from a) 1M 20mM, b) 2M 10mM and c) 2M 20mM, insets: EIS curves before and after polarization.

**Table S1)** The Li<sup>+</sup>-ion diffusion coefficient values of BF@QSSE coin-cells calculated from the CV measurements.

BF@QSSE	A1(cm <sup>2</sup> s <sup>-1</sup> )	A1(cm <sup>2</sup> s <sup>-1</sup> )	C1(cm <sup>2</sup> s <sup>-1</sup> )	C2(cm <sup>2</sup> s <sup>-1</sup> )
0.1 mV s <sup>-1</sup>	$5.63 \times 10^{-10}$	$9.38 \times 10^{-10}$	$3.45 \times 10^{-10}$	$6.35 \times 10^{-10}$
0.2 mV s <sup>-1</sup>	$5.52 \times 10^{-9}$	$1.22 \times 10^{-9}$	$4.11 \times 10^{-10}$	$5.81 \times 10^{-10}$
0.3 mV s <sup>-1</sup>	$6.45 \times 10^{-9}$	$1.2 \times 10^{-9}$	$4.31 \times 10^{-10}$	$5.18 \times 10^{-10}$
0.4 mV s <sup>-1</sup>	$6.86 \times 10^{-9}$	$1.23 \times 10^{-9}$	$4.60 \times 10^{-10}$	$4.79 \times 10^{-10}$



**Figure (S7).** a) Galvanostatic charge-discharge curves of BF@QSSE, BF@LE and PVDF@QSSE at 0.2 C. b) charge process of BF@QSSE, BF@LE and PVDF@QSSE cells showing the overpotentials for conversion between soluble LiPSs and insoluble Li<sub>2</sub>S<sub>2</sub>/Li<sub>2</sub>S.

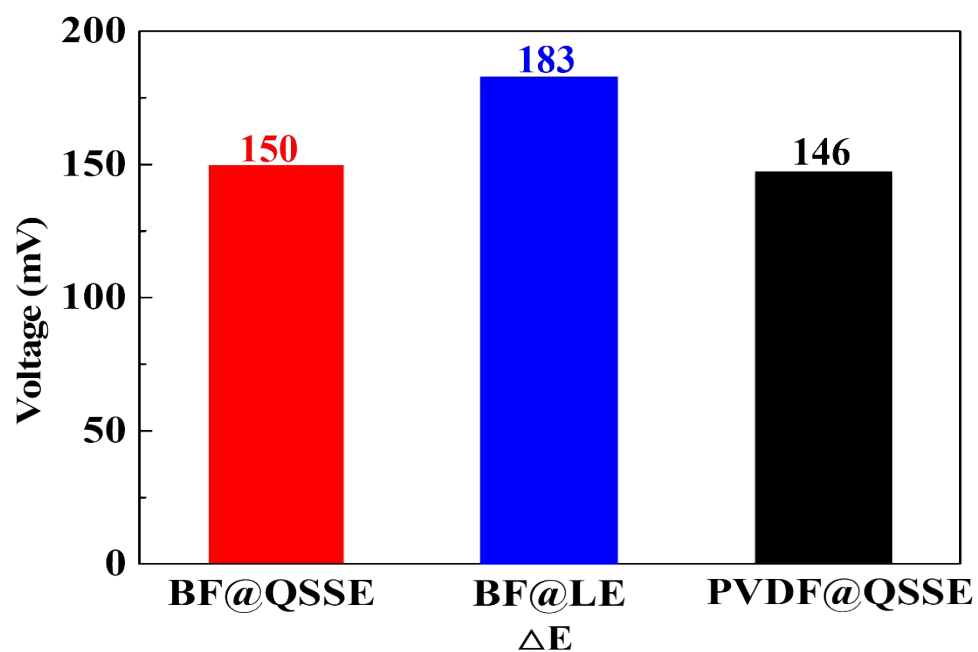


Figure (S8). The values of  $\Delta E$  obtained from charge–discharge voltage curves.

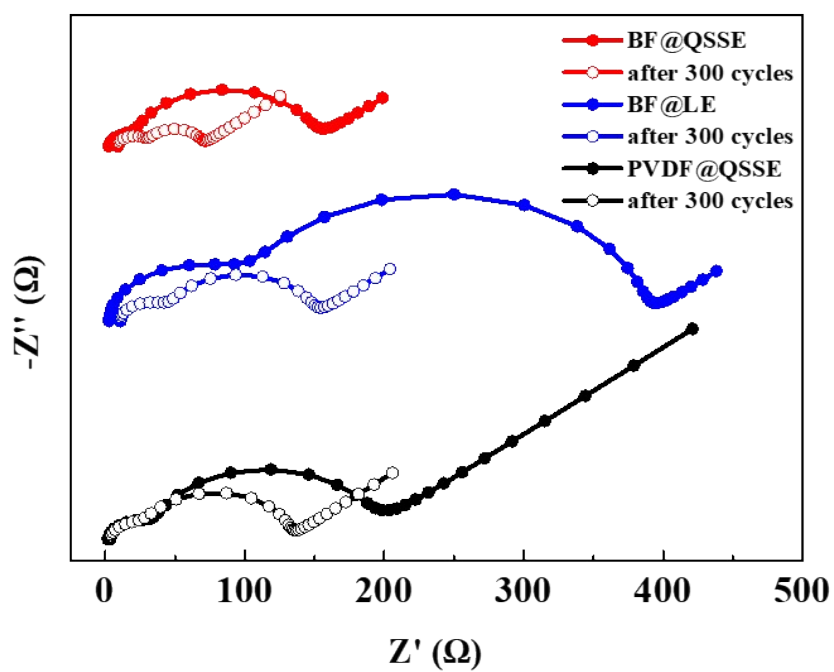


Figure (S9). Nyquist plots of BF@QSSE , BF@LE and PVDF@QSSE.

**Table S2)** EIS fitting parameters of BF@QSSE, BF@LE and PVDF@QSSE after 300 cycles at 1 C.

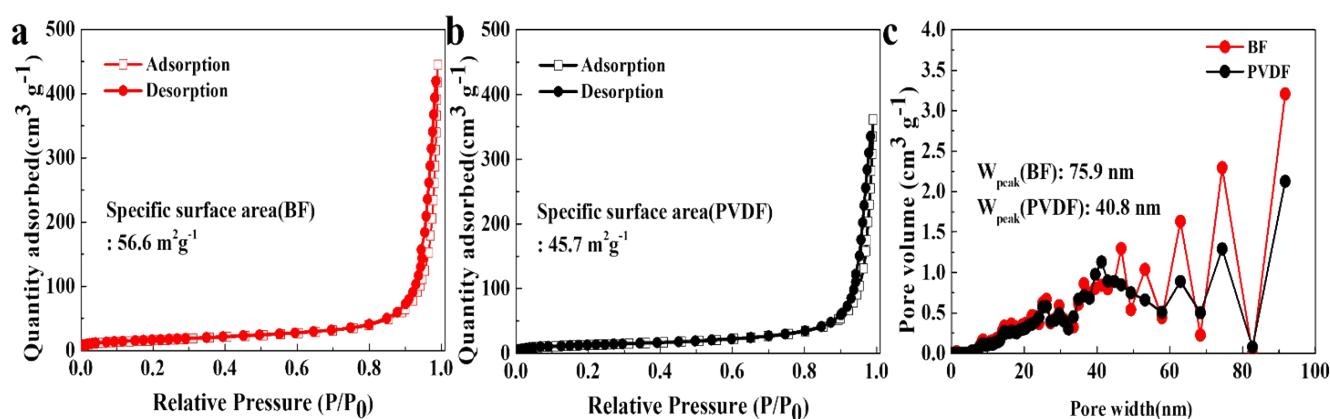
BF@QSSE	$R_b(\Omega)$	$R_{SEI}(\Omega)$	$R_{CT}(\Omega)$
Initial	2.48	18.17	122.64
After 300 cycles	9.12	21.40	36.73

BF@LE	$R_b(\Omega)$	$R_{SEI}(\Omega)$	$R_{CT}(\Omega)$
Initial	2.78	100.93	277.62
After 300 cycles	10.90	34.14	99.48

PVDF@QSSE	$R_b(\Omega)$	$R_{SEI}(\Omega)$	$R_{CT}(\Omega)$
Initial	1.97	31.32	146.64
After 300 cycles	3.48	24.35	101.88



**Figure (S10).**  $N_2$  adsorption–desorption isotherms of a) BF cathode and b) PVDF cathode c) pore size distribution of BF cathode and PVDF cathode.

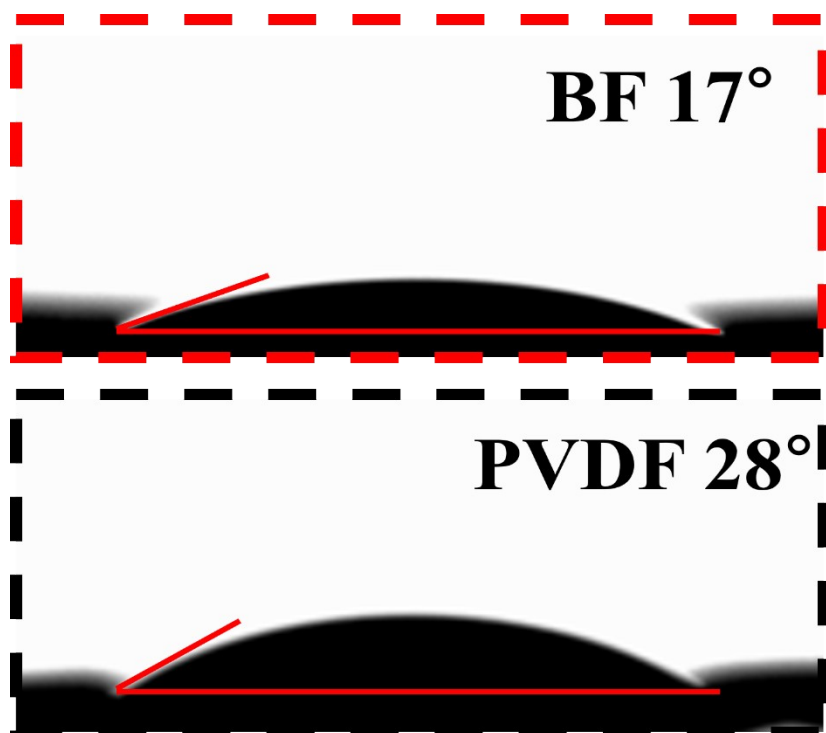


Figure (S11). Contact angle measurement of the BF cathode and PVDF cathode.

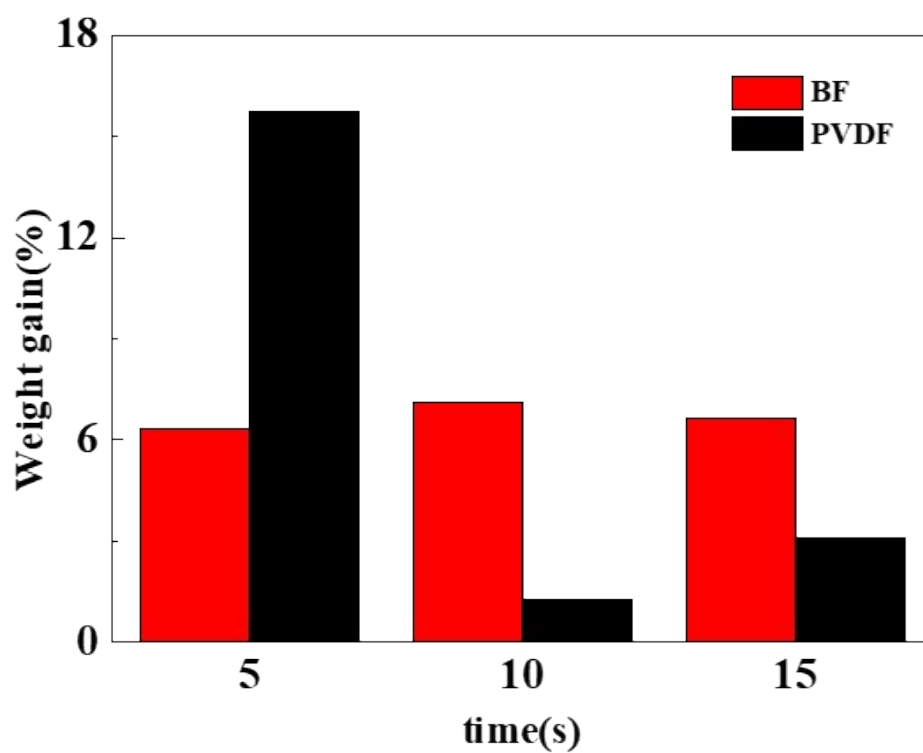
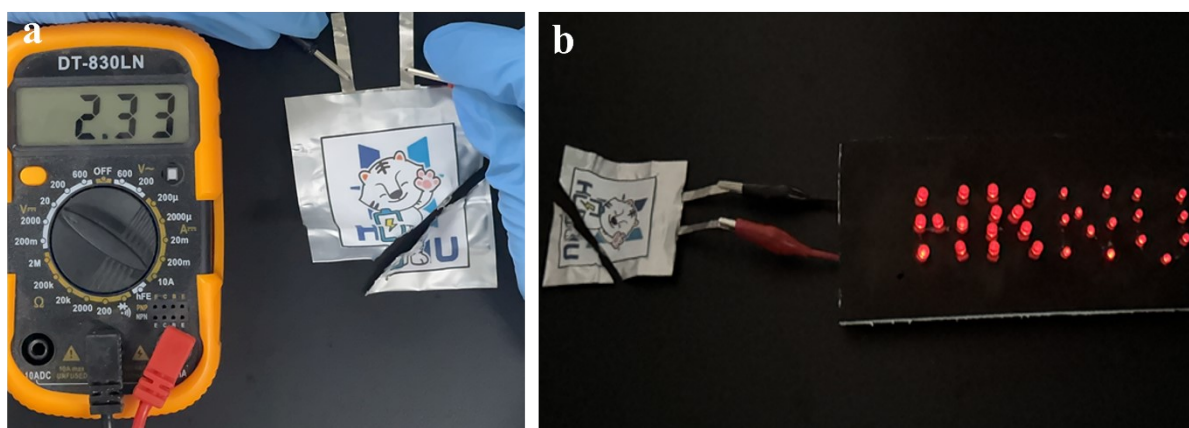


Figure (S12). Capillary effect of electrolyte absorption in BF and PVDF cathode.





**Figure (S13).** a) Voltage variation of the BF@QSSE pouch cell after cutting. b) Optical photograph of the flexible BF@QSSE pouch cell lighting up LEDs after cutting.