Supporting Information

Initiator loaded separator triggering in-situ polymerization of poly(1,3-dioxolane) quasi-solid electrolyte for lithium metal batteries

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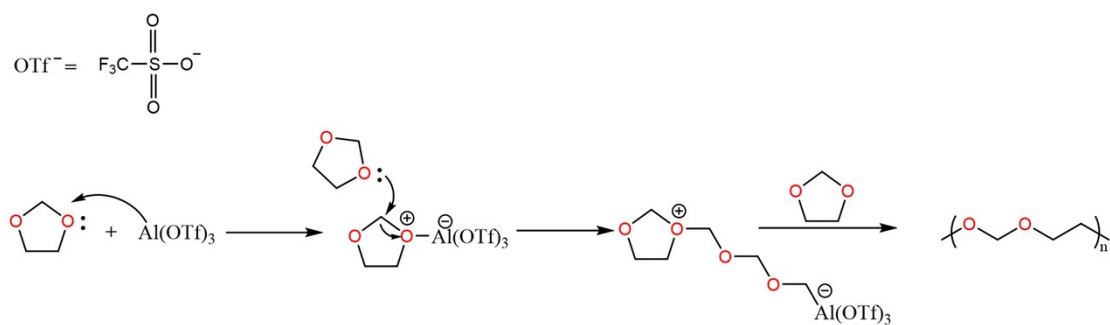Corresponding author E-mail: lijingze@uestc.edu.cn (J. Z. Li)

Fig. S1. Illustration of the reaction mechanism of DOL in situ polymerization.

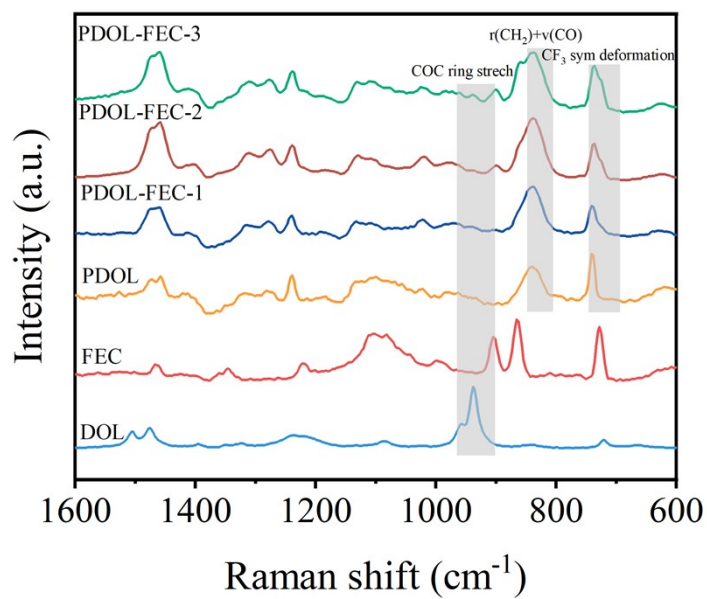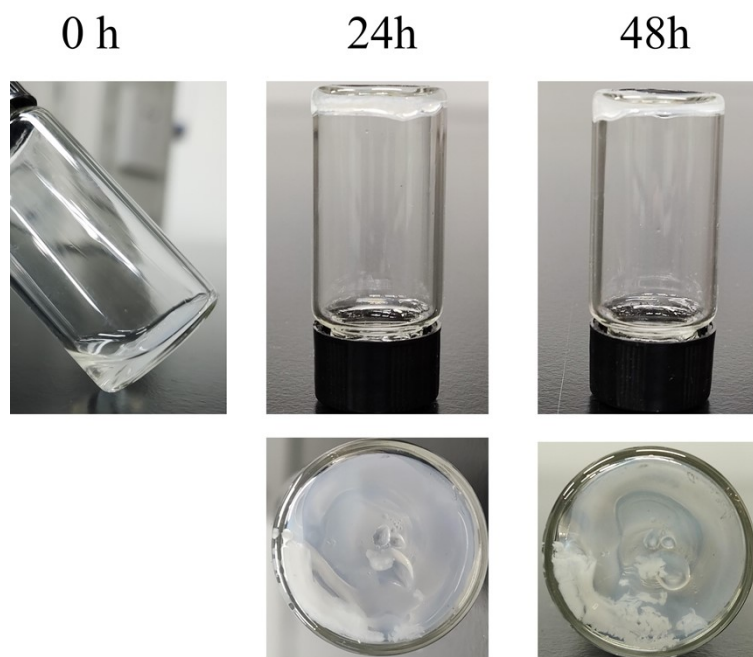
Fig. S2 Raman spectra of different electrolytes.

Fig. S3 Digital images of DOL precursor in the bottle before and after Al(OTf)3 loaded seperator initiated polymerization for 24 and 48h.

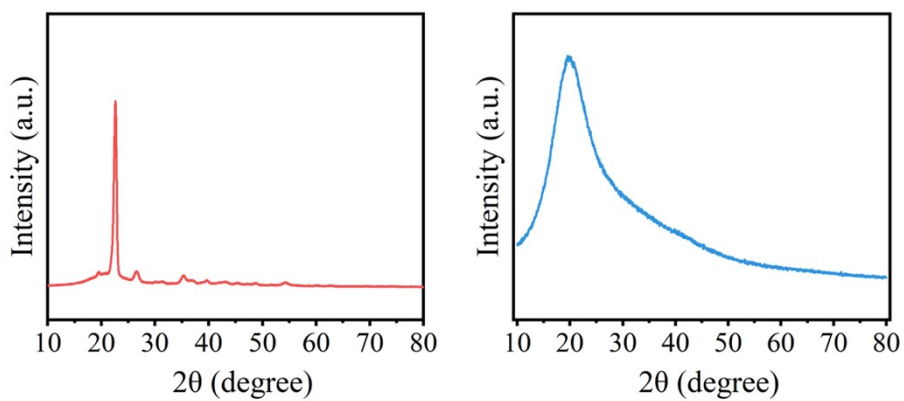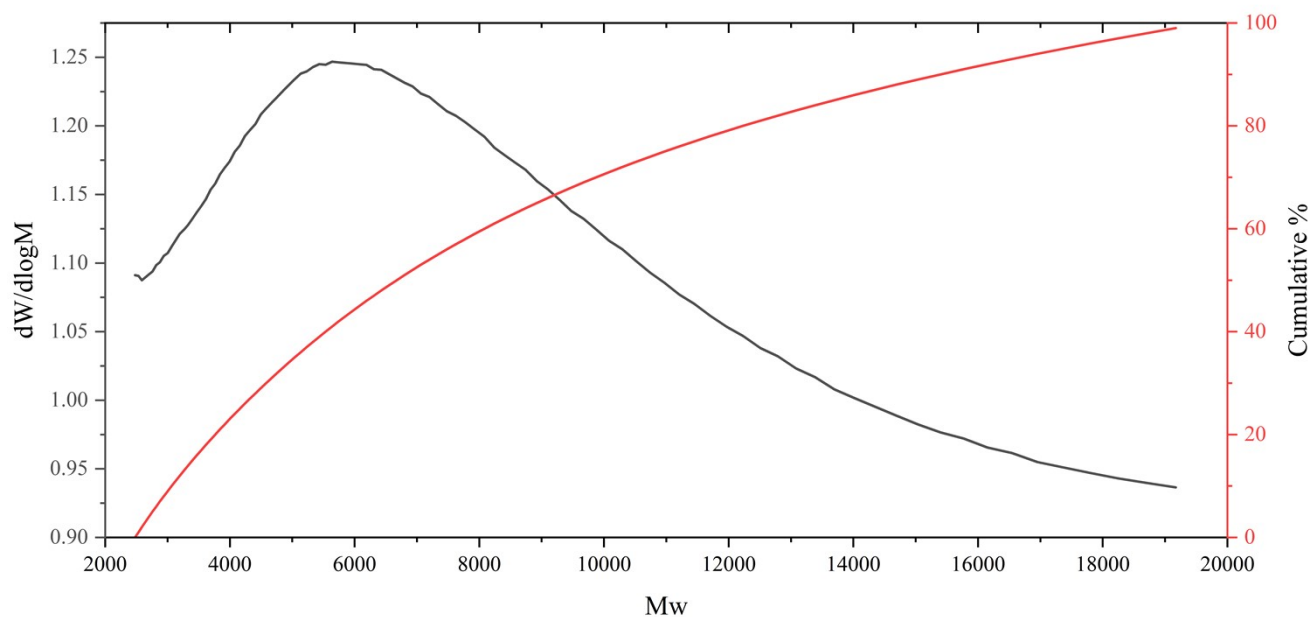Fig. S4 XRD patterns of different parts of the GPE. (a) near the separator and (b) away from the separator.

	Mn	Mw	Mw/Mn
PDOL-FEC-2	5739	7949	1.38

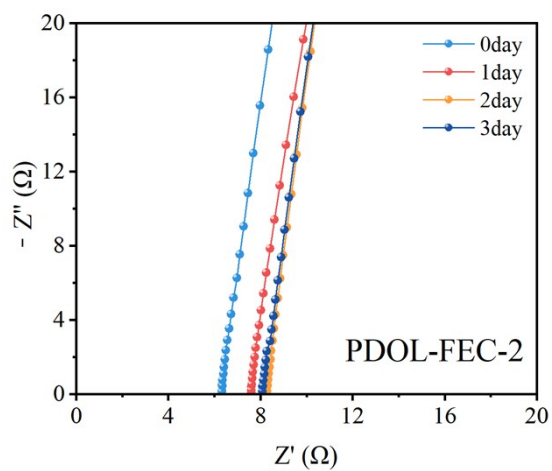Fig. S5. GPC data of PDOL-FEC-2 with THF as the solvent.

Fig. S6. EIS spectra of SS|PDOL-FEC-2|SS cell at room temperature as a function of the storage time.
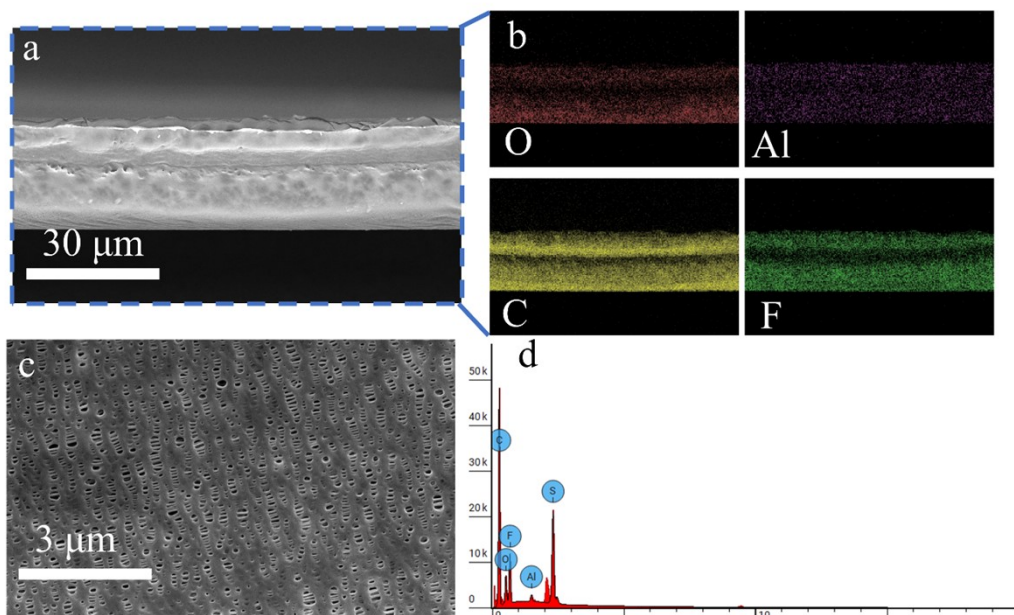
Fig. S7 (a) Cross-section SEM image of the Al (OTf)3-loaded separator. (b) Cross-section EDS mapping images of the cross-section of the Al (OTf)3-loaded separator. (c) Top-view SEM image of the PP separator. (d) EDS curve of the composite separator.
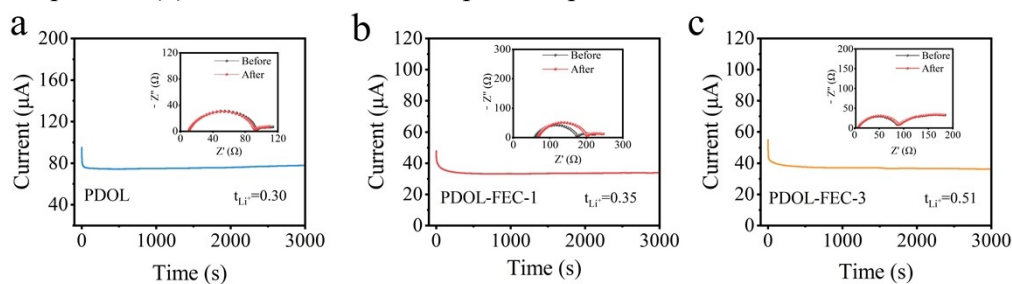

Fig. S8 chronoamperometry curves with a step voltage of 10 mV of cells with (a)PDOL, (b)PDOL-FEC-1, and (c)PDOL-FEC-3.The insets displayed EIS plots before and after polarization.
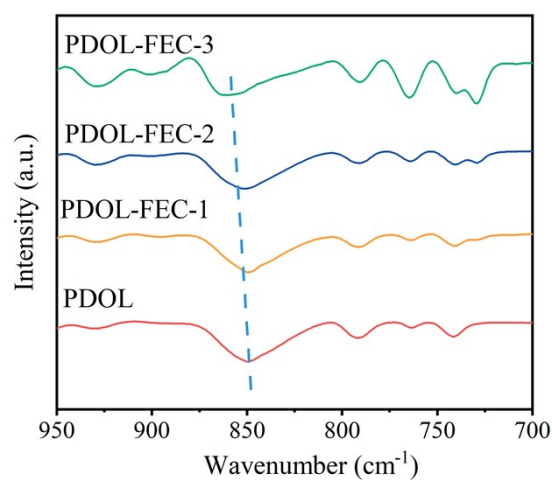

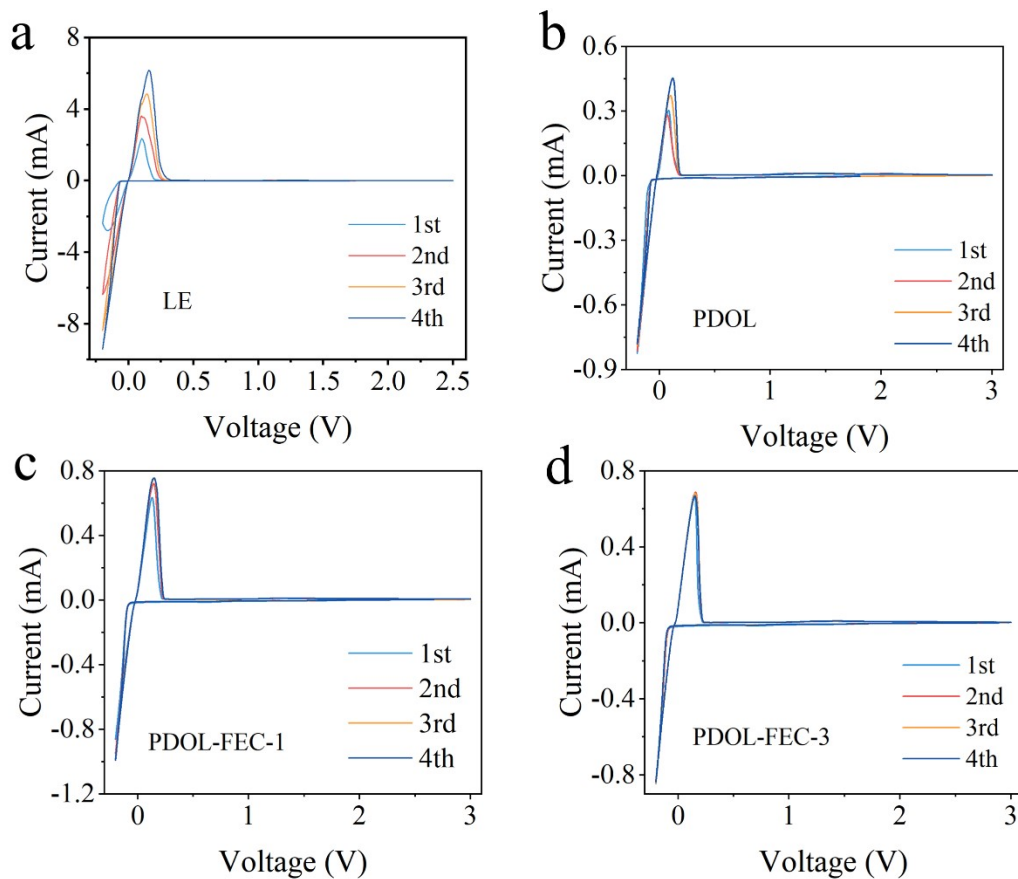Fig. S9. FTIR spectra of PDOL, PDOL-FEC-1, PDOL-FEC-2, and PDOL-FEC-3.

Fig. S10. CV curves of the Li||SS cells with (a)LE, (b)PDOL, (c)PDOL-FEC-1, and (d)PDOL-FEC-3.
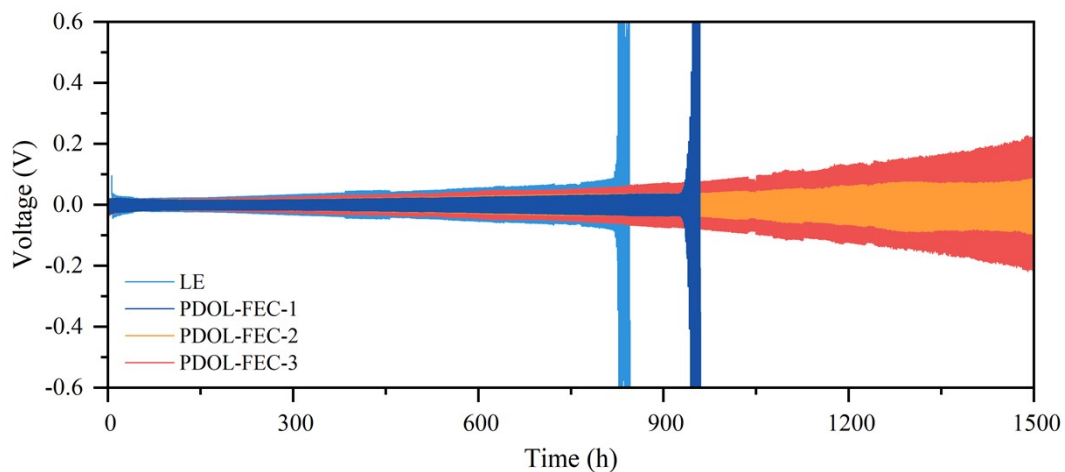
Fig. S11. Voltage profiles of Li//Li symmetrical cells that contain different electrolytes at room temperature with the current density of 0.5 mAh cm^{-2}.

Fig. S12. Voltage profiles of Li‖Li cells at different current densities.

Fig S13 XPS spectra (C 1s, F 1s and Li 1s spectra) of the Li metal surface from the cycled Li‖Li cells after 50 h at 1 mA cm^{-2} for 1 mA h cm^{-2}: (a，b and c) LE cell and (d, e and f) GPE cell (PDOL-FEC-2)
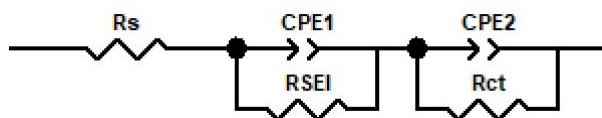
Fig. S14 Equivalent circuit for fitting Nyquist plots of Li‖Li symmetric cells.
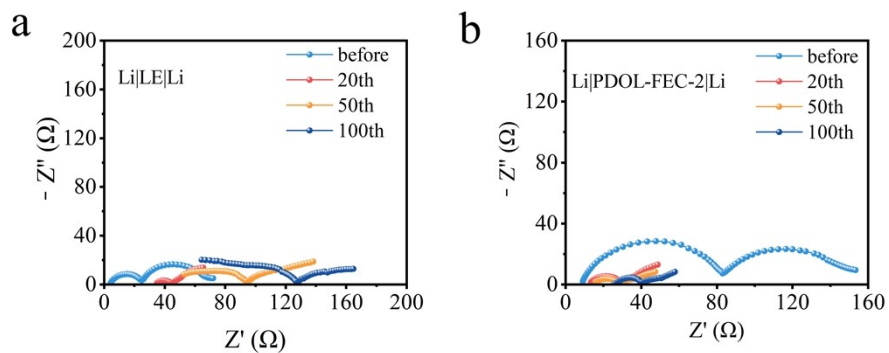
Fig. S15. The EIS of Li/Li cells with the LE (a) and (b) PDOL-FEC-2.

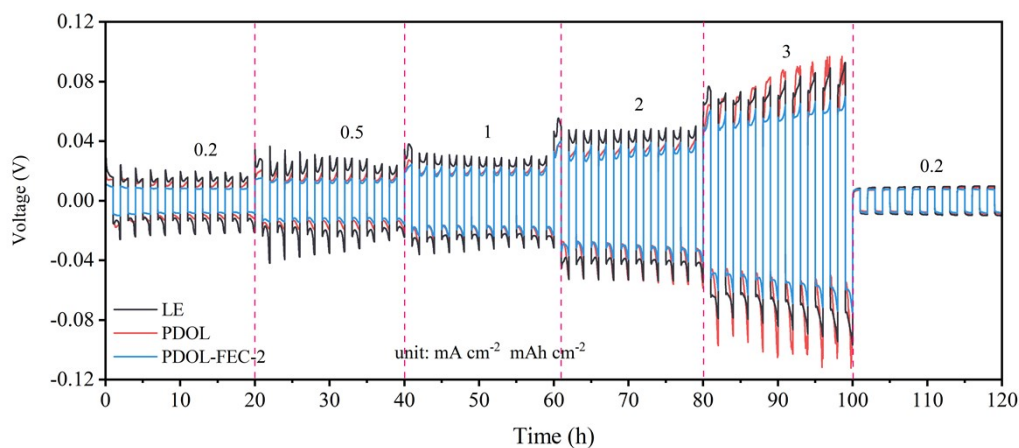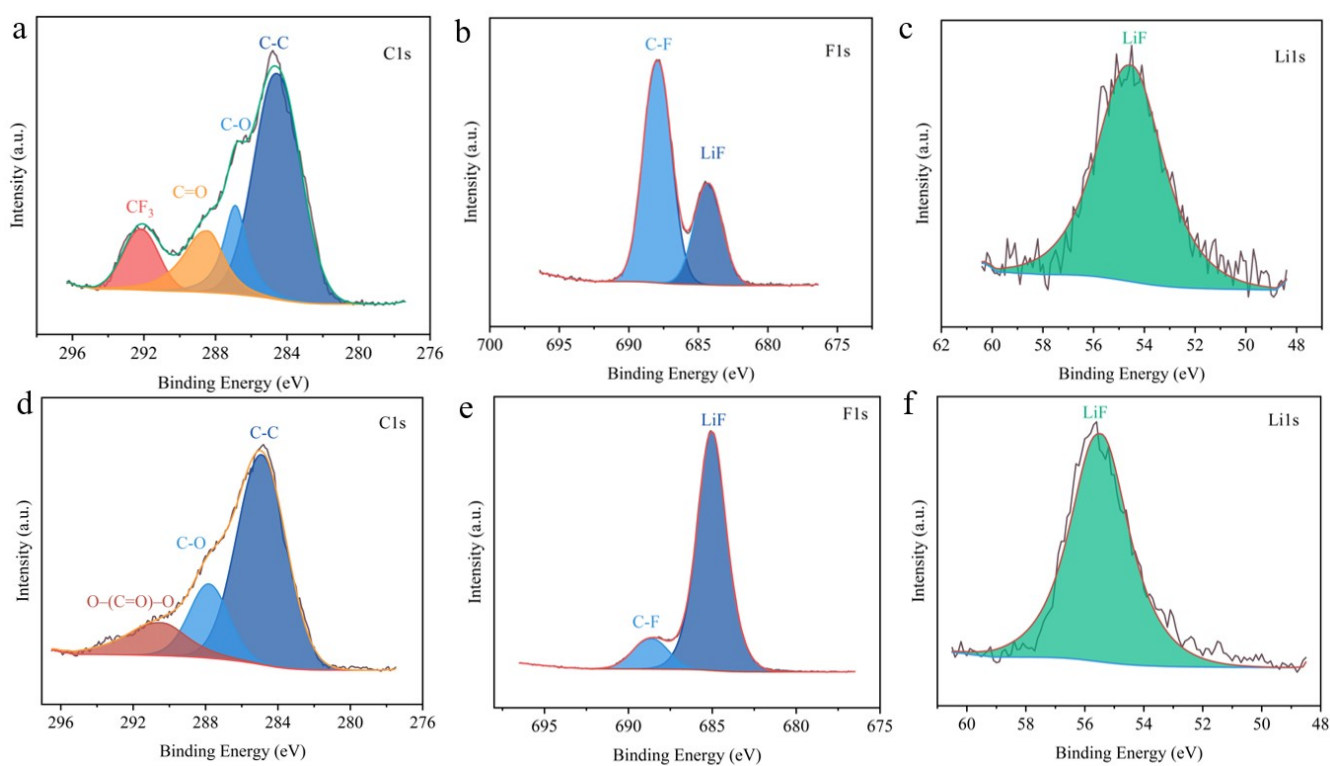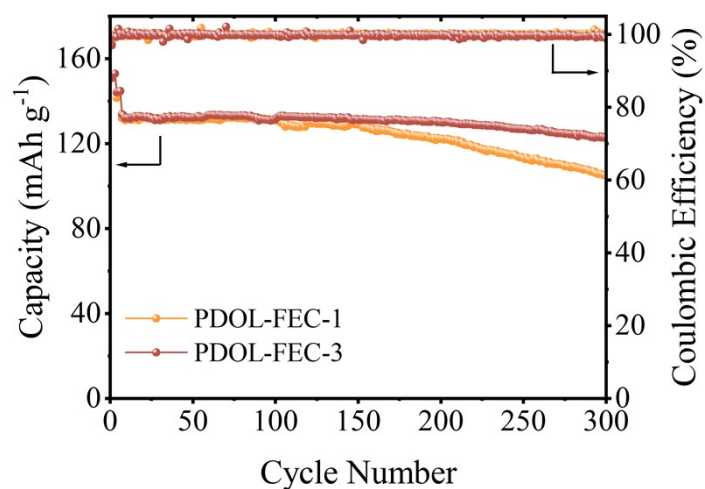Fig. S16. Cycling performances and CE of the LFP||Li cell at room temperature with PDOL-FEC-1 and PDOL-FEC-3 electrolytes at 1 C.

Table S1 Liquid content of different electrolytes

Sample	liquid content.
PDOL	2.2%
PDOL-FEC-1	4.5%
PDOL-FEC-2	9.2%
PDOL-FEC-3	10.2%

Table S2 The fitted EIS parameters of Li|LE|Li and Li|PDOL-FEC-2|Li batteries

		R_S	R_{SEI}	R_{ct}
Li|LE|Li	Before	4.238	19.72	46.35
	20th	9.911	6.362	4.229

	50th	13.81	8.716	5.186
	100th	28.47	12.36	6.782
Li|PDOL-FEC-2|Li	Before	8.459	75.08	61.84
	20th	12.63	16.66	16.25
	50th	16.14	11.55	16.75
	100th	27.41	12.53	17.71

Table S3 Comparison of our work with previously reported PDOL-based electrolytes via in-situ polymerization

Electrolyte	Initiator	Ionic Conductivity, t_{Li}^+	Li//Li cell performance	Battery performance	Ref
2 M LiTFSI +DOL	0.5mM Al (OTf)$_3$	1×10^{-3} S cm^{-1}	400 h; 1.0 mA cm^{-2}, 1.0 mAh cm^{-2}	1 C, 700 cycles	[1]
DOL+DME +1M LiTFSI	2M LiPF$_6$	5.50×10^{-4} S cm^{-1}	400 h; 1.0 mA cm^{-2}, 1.0 mAh cm^{-2}	0.5 C, 700 cycles, capacity retention:95.6%	[2]
DOL/FEC (v/v=10:1) +2M LiTFSI	0.43mM Al(OTf)$_3$	2.4×10^{-5} S cm^{-1} at -60 °C; 0.55 at -20°C	850 h; 0.2 mA cm^{-2}, 0.2 mAh cm^{-2} at 0°C	0.2 C, 400 cycles, capacity retention:96% at 0 °C	[3]
2 M LiTFSI +DOL	Sc(OTf)$_3$	1.07×10^{-3} S cm^{-1}	/	1 C, 200 cycles, capacity retention:92.3%	[4]
2 M LiTFSI +DOL	0.4mM Al (OTf)$_3$	2.39×10^{-3} S cm^{-1}	250 h; 0.5 mA cm^{-2}, 1.0 mAh cm^{-2}	1 C, 200 cycles, capacity retention:69.96%	[5]
DOL+DME+1M LiTFSI	acid-treated nano Al$_2$O$_3$	3.37×10^{-3} S cm^{-1}; 0.74	1000 h; 1.0 mA cm^{-2}, 1.0 mAh cm^{-2}	200cycles, capacity retention:82.1%	[6]
DOL+SCN+FEC +1 M LiTFSI	0.4 M LiBF$_4$	1.95×10^{-4} S cm^{-1}; 0.36	600 h; 0.2 mA cm^{-2}, 0.2 mAh cm^{-2}	0.5C,1000 cycles, capacity retention:80%	[7]
DOL/FEC (v/v=10:2) +1M LiTFSI	Al (OTf)$_3$	1.5×10^{-4} S cm^{-1}; 0.63	650 h; 1.0 mA cm^{-2}, 1.0 mAh cm^{-2}	1C, 300 cycles, capacity retention:94.5%	This work

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