

ARTICLE

## In-situ polymerization of electrochemically stable dual-salt gel polymer electrolyte for lithium ion batteries† (Electronic Supplementary Information)

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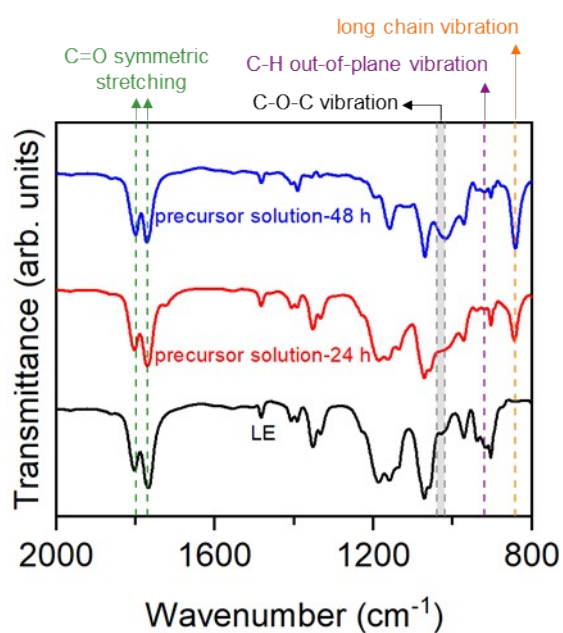


Fig. S1 FTIR spectrum of precursor solution with different gelation times and LE.

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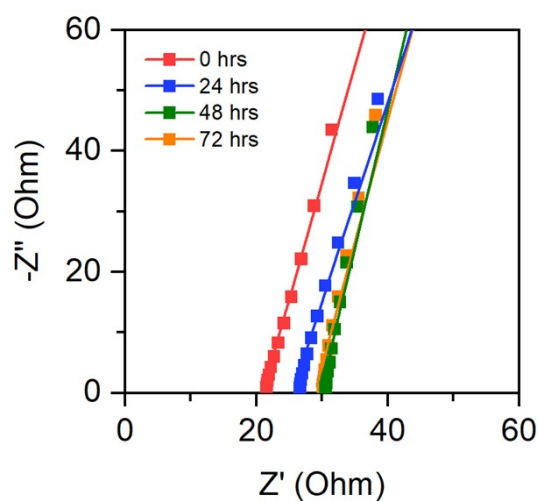


Fig. S2 EIS spectra of GPE with different gelation times.

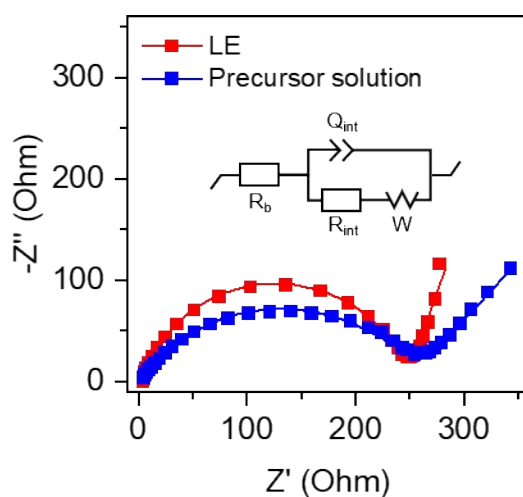


Fig. S3 EIS spectra of Li | precursor solution | LFP and Li | LE | LFP cells. (Inset is the equivalent circuit to fit the plots, where  $R_b$  represents the equivalent series resistance,  $R_{int}$  represents the interfacial resistance between electrolyte and electrode, and  $W$  represents Warburg impedance.)

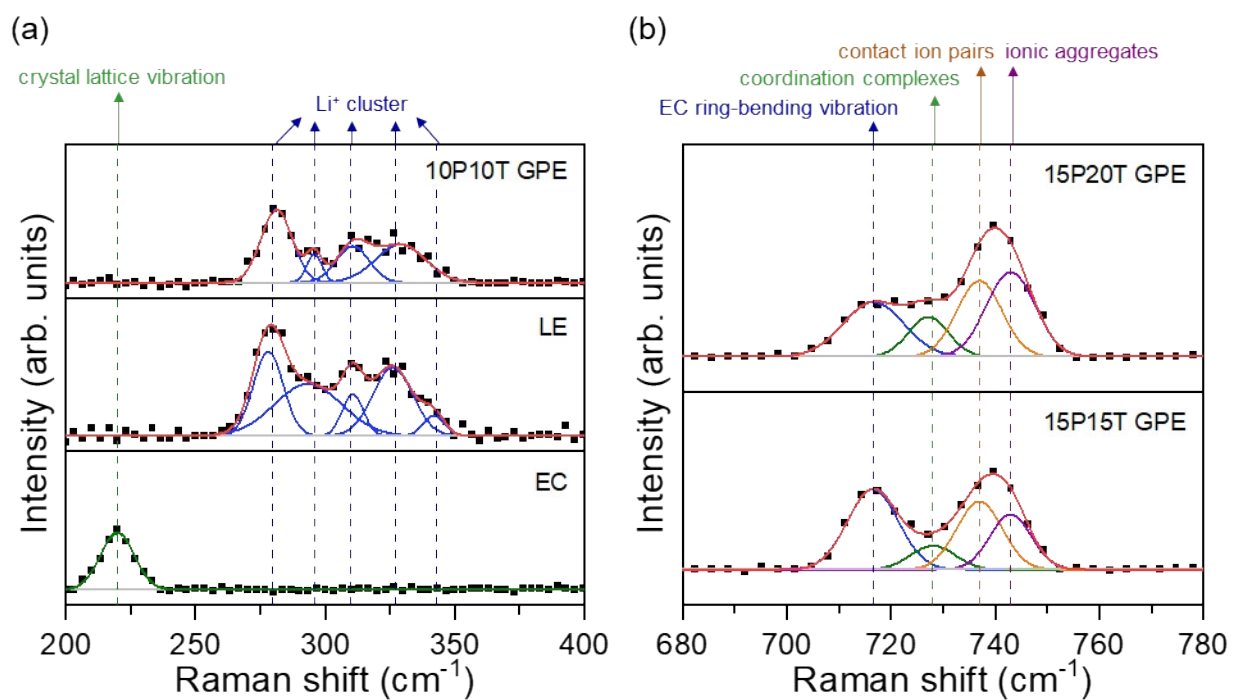


Fig. S4 Raman spectra of (a) EC, LE and 10P10T GPE. (b) 15P15T and 15P20T GPE. (square dots: experiment points; solid lines: fitting results)

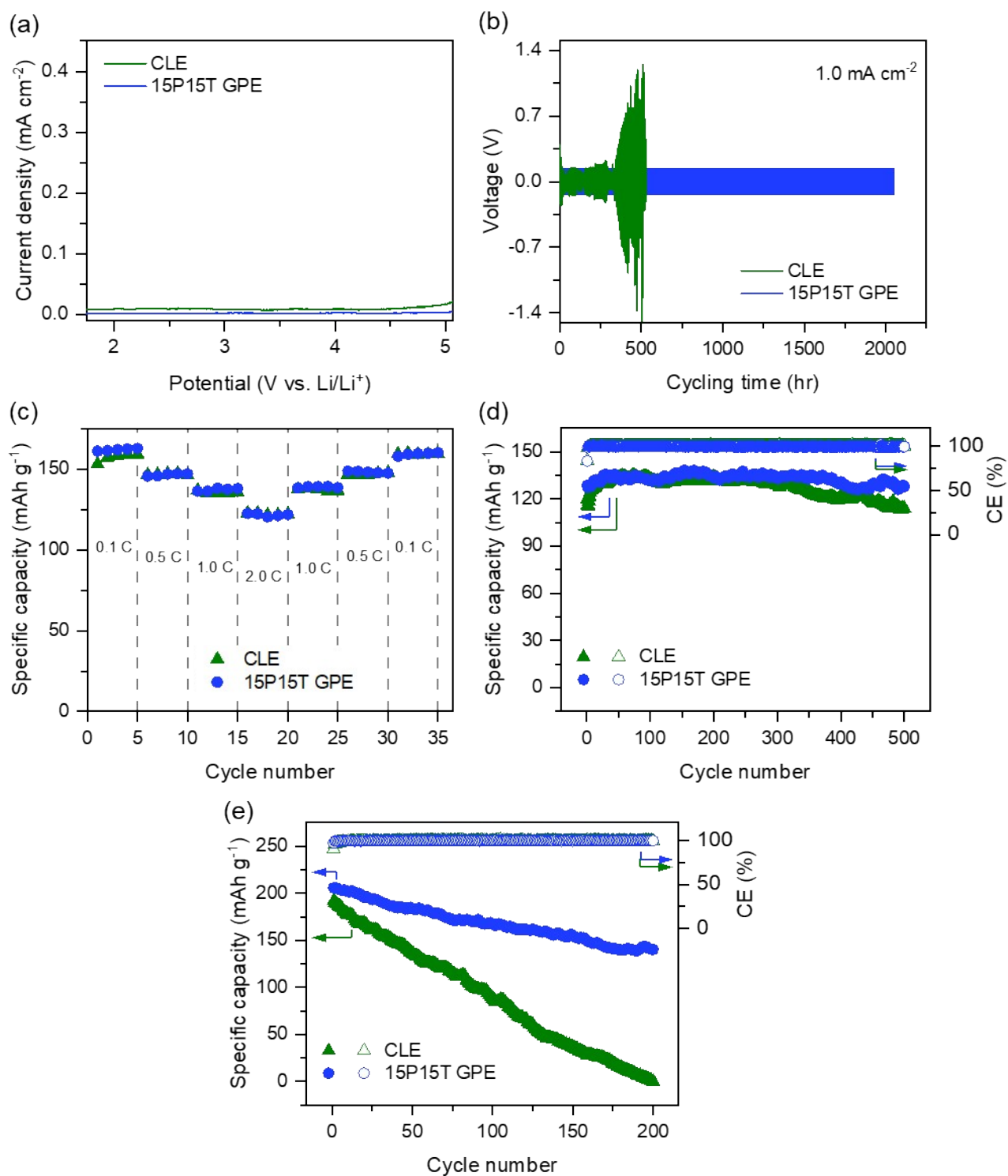


Fig. S5 (a) LSV curves of CLE and 15P15T GPE. (b) Cyclic lithium stripping/plating behaviors of Li||Li symmetrical batteries with CLE and 15P15T GPE. (c) Rate performance of Li||CLE||LFP and Li||15P15T GPE||LFP cells. (d) Cycle performance and Coulombic efficiencies of Li||CLE||LFP and Li||15P15T GPE||LFP cells at 1 C. (e) Cycle performance and Coulombic efficiencies of Li||CLE||NCM811 and Li||15P15T GPE||NCM811 cells at 0.2 C.

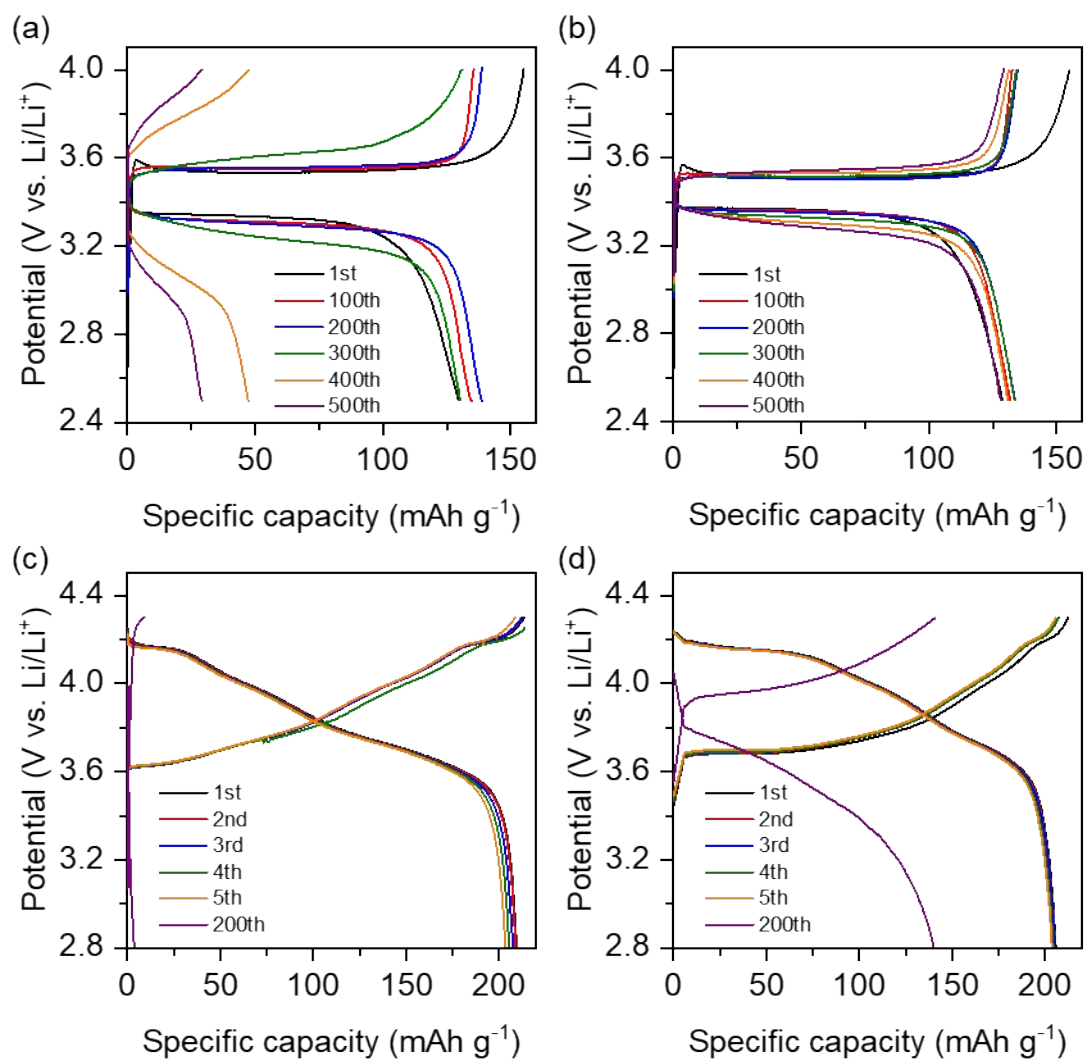


Fig. S6 Charge/discharge curves of (a) Li||LE||LFP and (b) Li||15P15T GPE||LFP cells at 1 C. Charge/discharge curves of (c) Li||LE||NCM811 and (d) Li||15P15T GPE||NCM811 cells at 0.2 C.

Table S1 The best fitting results of the interfacial resistance for the impedance spectra of Li||precursor solution||LFP and Li||LE||LFP cells.

Sample	Bulk resistance (ohm)
Precursor solution	251.0
LE	243.7

Table S2 Results of degree of polymerization, ionic conductivity, and Li<sup>+</sup> transference number of LE and different LiPF<sub>6</sub> concentrations of GPE.

Sample	Degree of polymerization	Bulk resistance (ohm)	Ionic conductivity (S/cm)	Li <sup>+</sup> transference number
LE	-	1.7	$7.30 \times 10^{-4}$	0.25
10P10T GPE	78.9%	45.0	$2.76 \times 10^{-5}$	0.42
15P10T GPE	80.5%	37.0	$3.35 \times 10^{-5}$	0.57
20P10T GPE	78.0%	44.1	$2.81 \times 10^{-5}$	0.50

Table S3 Results of degree of polymerization, ionic conductivity, and Li<sup>+</sup> transference number of different LiTFSI concentrations of GPE.

Sample	Degree of polymerization	Bulk resistance (ohm)	Ionic conductivity (S/cm)	Li <sup>+</sup> transference number
15P10T GPE	80.5%	37.0	$3.35 \times 10^{-5}$	0.57
15P15T GPE	84.3%	29.8	$4.16 \times 10^{-5}$	0.63
15P20T GPE	77.6%	39.3	$3.15 \times 10^{-5}$	0.56

Table S4 The proportion of peak areas in the Raman spectra of the 15P15T and 15P20T GPE.

Sample	Coordination complexes	Contact ion pairs	Ionic aggregates
15P15T GPE	16.4%	48.1%	35.5%
15P20T GPE	17.5%	37.5%	45.0%