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## Supporting Information

### Enhanced Ion Transport Behaviors in Composite Polymer Electrolyte: the Case of Looser Chain Folding Structure

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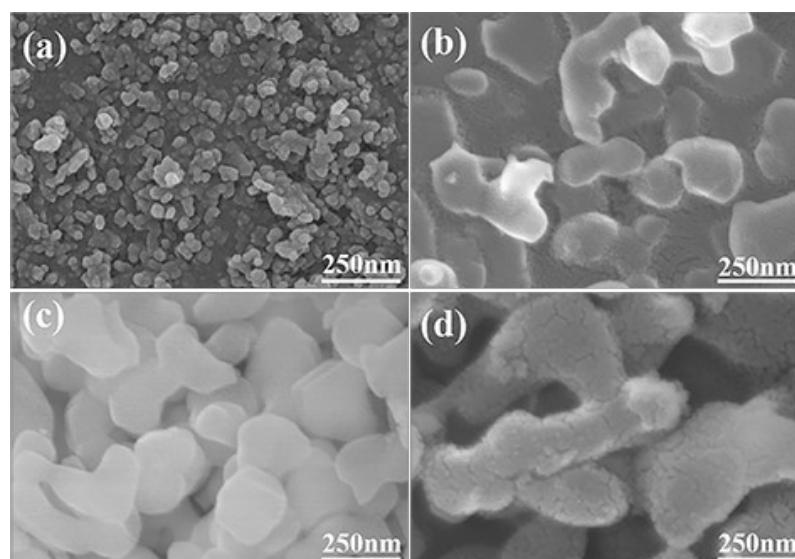


Figure S1. SEM images of Al<sub>2</sub>O<sub>3</sub> particles of (a) 30 nm, (b) 100 nm, (c) 200 nm, and (d) 400 nm

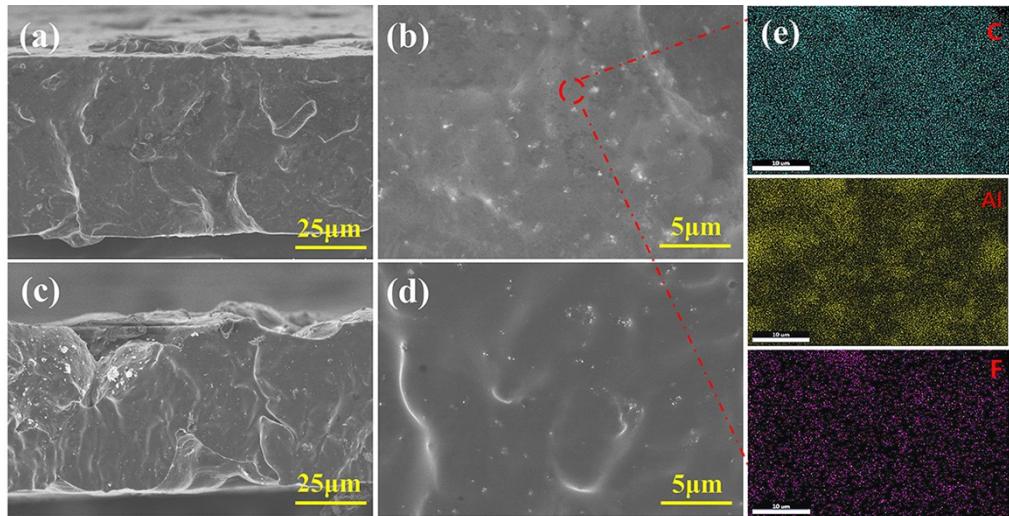


Figure S2. (a, b) Cross-section SEM images of CPEs-30, (c, d) CPEs-200, and (e) EDS mapping of CPEs-30

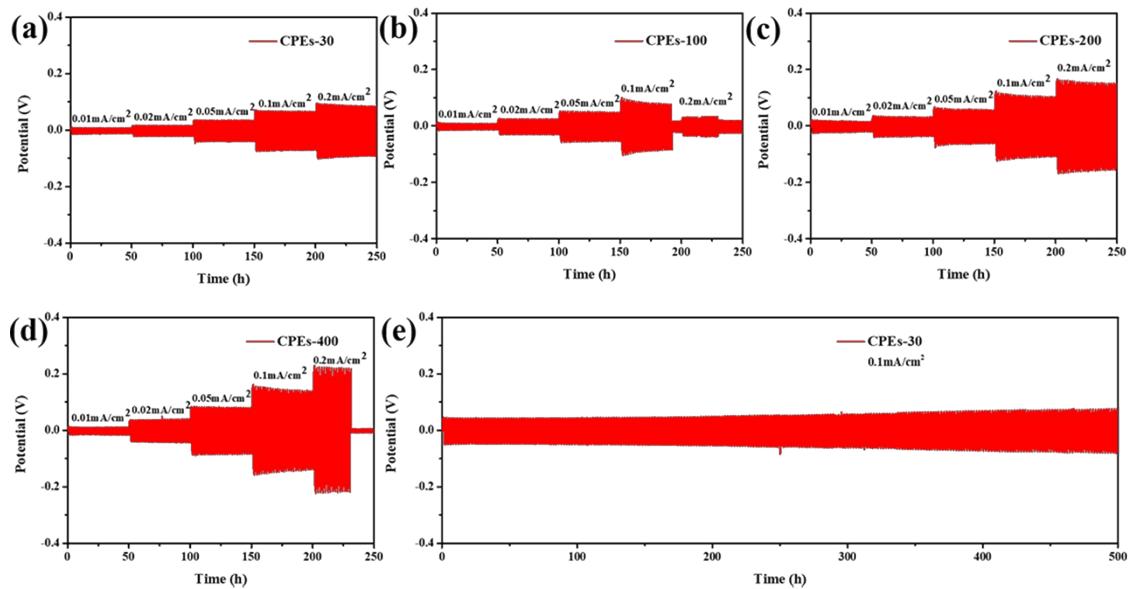


Figure S3. Voltage profile of the lithium plating/stripping cycling in the symmetrical (a) Li/CPEs-30/Li cell, and (b) Li/CPEs-100/Li cell, (c) Li/CPEs-200/Li cell, (d) Li/CPEs-400/Li cell with different current densities, and (e) plating/stripping cycling at 0.1 mA/cm<sup>2</sup> of CPEs-30

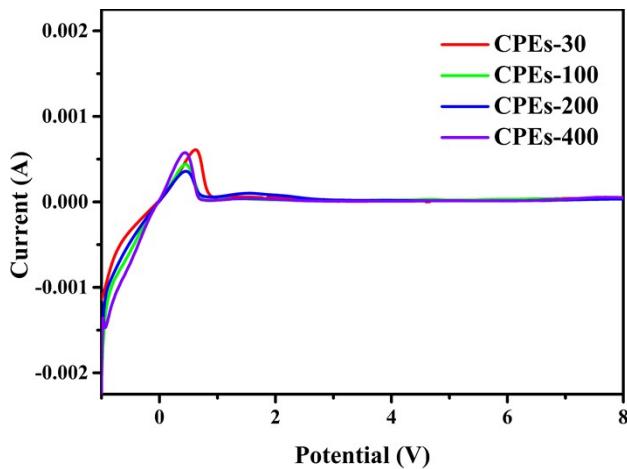


Figure S4. The LSV of CPEs-30, CPEs-100, CPEs-200, and CPEs-400 at  $0.1 \text{ mV s}^{-1}$  from  $-1 \text{ V} \sim 8 \text{ V}$  in Li//CPEs//Steel cells

Table S1. Electrochemical performances of CPEs with different kinds of ceramic fillers

Electrolytes	Cathode/anode	Capacity(mAh/g)	Cycle Performance	Year	REF
CPEs-30	LFP/Li	118.2 (1 C)	81.3% (1 C, 500 cycles)	This work	
PVDF-HFP-EC-DC/LiPF <sub>6</sub> -Al <sub>2</sub> O <sub>3</sub>	LFP/Li	155 (0.5 C)	95.6% (0.5 C, 100 cycles)	2018	1
PPC-LiTFSI-Al <sub>2</sub> O <sub>3</sub>	NCM622/Li	168.9 (0.5 C)	90.9% (0.5 C, 100 cycles)	2021	2
PEO-LiClO <sub>4</sub> -SiO <sub>2</sub>	LFP/Li	120 (1 C)	87.5% (1 C, 60 cycles)	2016	3
PEC-FEC-PTFE-LiFSI-LiMNT	LFP/Li	137.5 (0.5 C)	91.9% (0.5 C, 200 cycles)	2019	4
PVDF-HFP-PEG-Al <sub>2</sub> O <sub>3</sub>	LFP/Li	132.1 (1 C)	78.8% (1 C, 500 cycles)	2021	5
PVDF-HFP-LiClO <sub>4</sub> -LLZO	LFP/Li	120 (0.5 C)	92.5% (0.5 C, 180 cycles)	2018	6
PVDF/PAN/LiTFSI-SN-LiNf@Ga/F-LLZO	LMO@T-LNCM811/Li	112.8 (1 C)	89.8% (1 C, 300 cycles)	2021	7
PEO-LiTFSI-LLZO	LFP/Li	159.3 (0.2 C)	84% (0.2 C, 450 cycles)	2021	8
PEO-LiTFSI-LLZO	LFP/Li	159.9 (0.5 C)	97.4% (0.5 C, 70 cycles)	2019	9
PEO-LiClO <sub>4</sub> -LLZTO	LFP/Li	140 (1 C)	83% (1 C, 500 cycles)	2017	10
PVDF-HFP-LiTFSI-	LFP/Li	158.2 (0.5 C)	93.2% (0.5 C, 150 cycles)	2021	11

LATP

PEO-PVDF-HFP-LITFSI-LATP	LFP/Li	113.1(0.8 C)	86.7% (0.8 C, 500 cycles)	2020	12
PCL-LITFSI-LAGP	LFP/Li	157 (0.1 C)	96% (0.1 C, 130 cycles)	2021	13
PEO-LITFSI-LAGP	LFP/Li	148.7 (0.3 C)	93.3 (0.3 C, 300 cycles)	2019	14

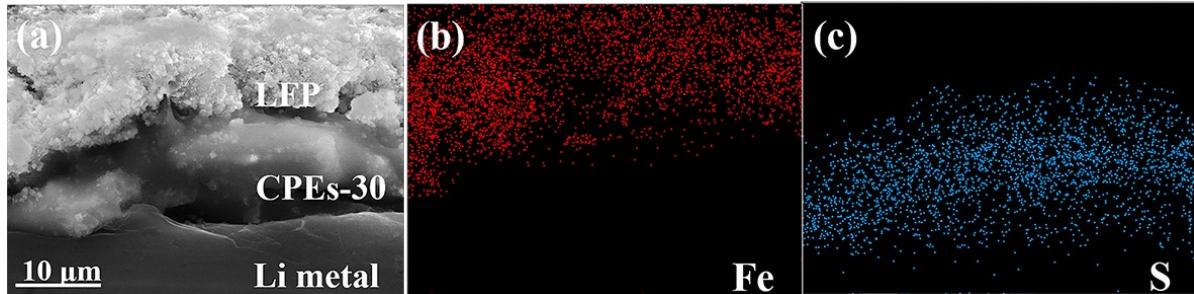


Figure S5. SEM images of cross-section of (a) LFP/CPEs-30/Li cell, and (b, c) EDS mapping of the cross-section

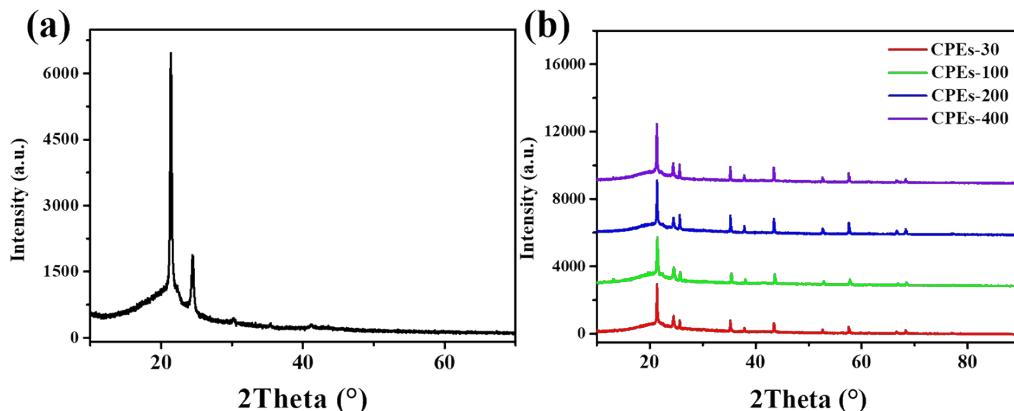


Figure S6. (a) XRD patterns of the pure PCL, (b) XRD patterns of CPEs-30, CPEs-100, CPEs-200, and CPEs-400

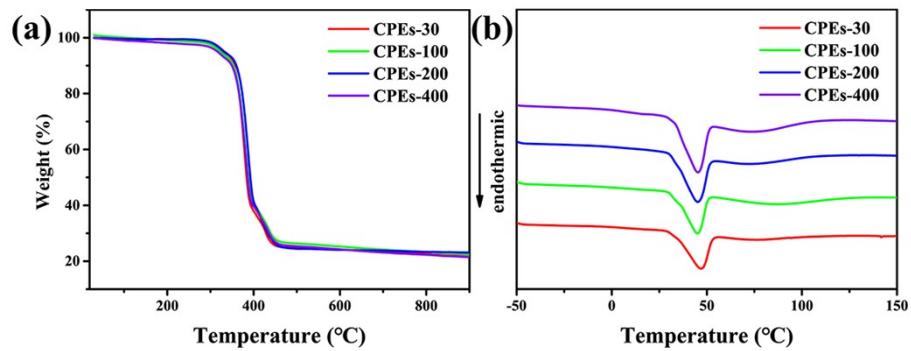


Figure S7. (a) TGA curves of CPEs-30, CPEs-100, CPEs-200, and CPEs-400, and (b) DSC curves of CPEs-30, CPEs-100, CPEs-200, and CPEs-400

Table S2. Thermal properties ( $T_g$ ,  $T_m$ , and  $\Delta H_m$ ) and crystalline ( $\chi_c$ ) of different CPEs obtained from DSC results

Composite polymer electrolyte	$T_g$ (°C)	$T_m$ (°C)	$\Delta H_m$ (Jg <sup>-1</sup> )	$\chi_c$ (%)
CPEs-30	-49.21	46.71	40.84	40.72
CPEs-100	-48.64	44.85	41.47	41.35
CPEs-200	-51.20	45.01	45.44	45.31
CPEs-400	-52.13	45.14	47.13	47.00

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