

In-Situ Constructing of Ultra-Thin and Flexible Polymer Electrolyte for Stable All-Solid-State Lithium-Metal Batteries

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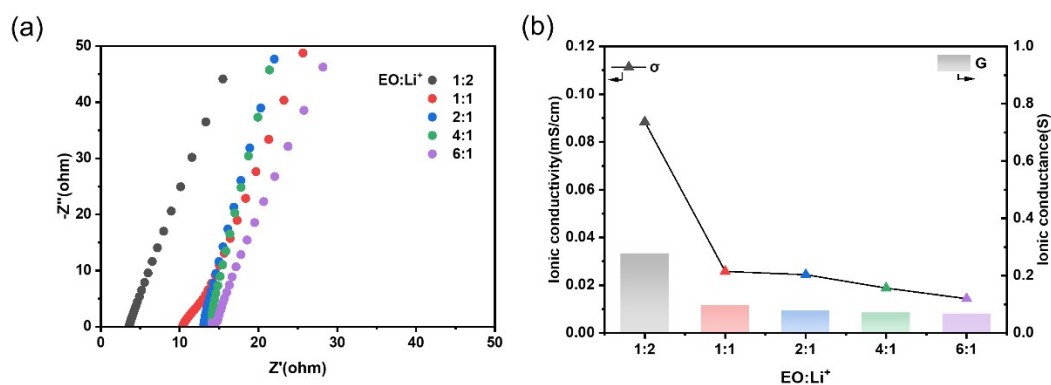


Figure S1. (a) The EIS curves and (b) corresponding ionic conductivity and conductance of the polymer electrolyte with different molar ratio of EO:Li⁺.

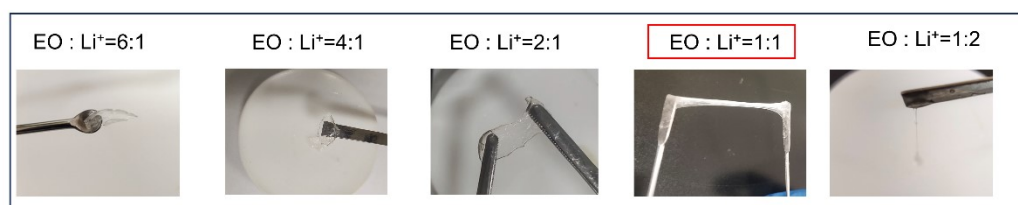


Figure S2. The digital photos of the polymer electrolyte with different molar ratios of EO:Li⁺.

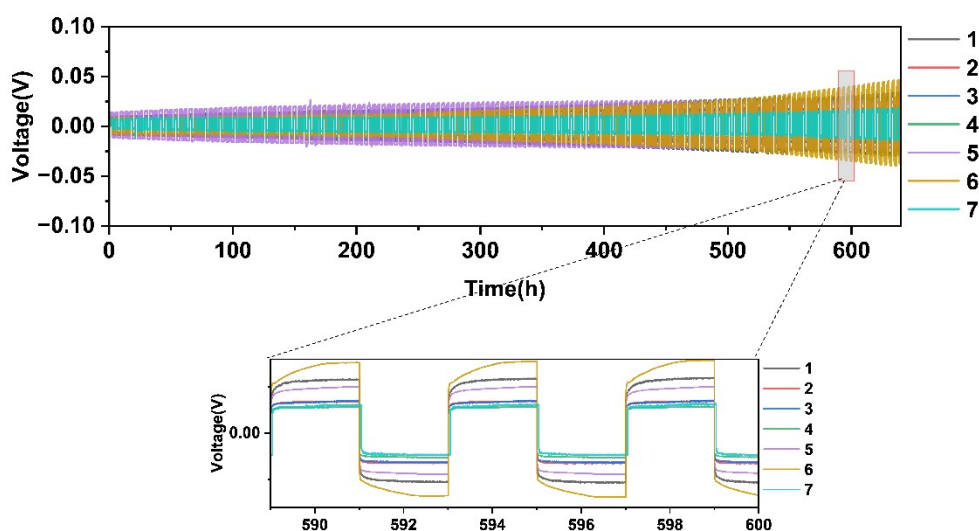


Figure S3. Voltage profiles of the Li||Li cells at current density of 0.1 mA cm^{-2} with electrolytes prepared by different ratio of monomers (for example: electrolyte 1 was prepared by PEGMEMA:VEC:DMAPMA=0.5:1:1, see Table S2 for more details)

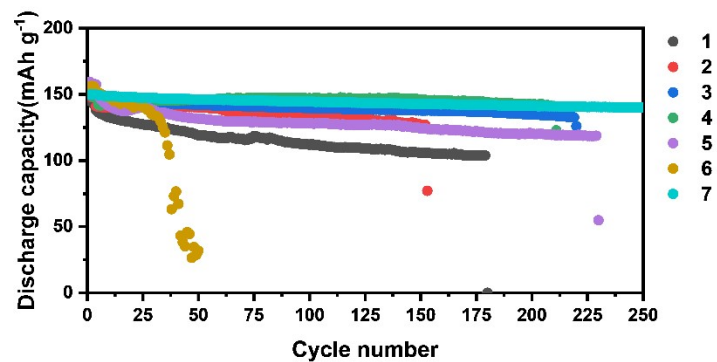


Figure S4. Cycling performance of the Li||LiFePO₄ cells at current density of 0.5 C with different ratios of monomers

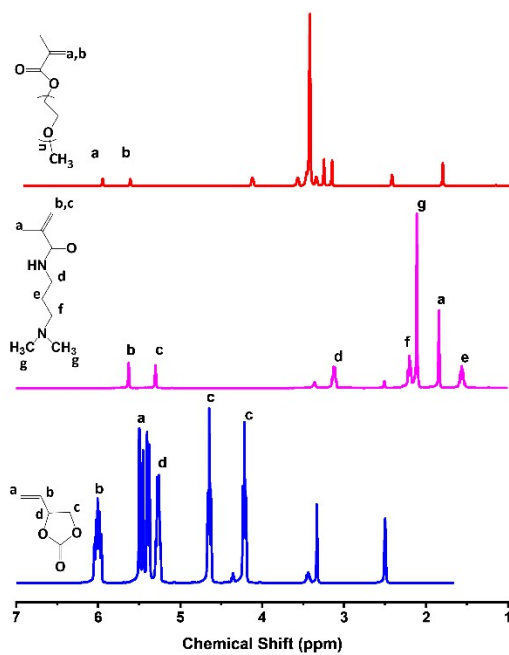


Figure S5. ¹H NMR spectra of PEGMEMA, DMAPMA and VEC.

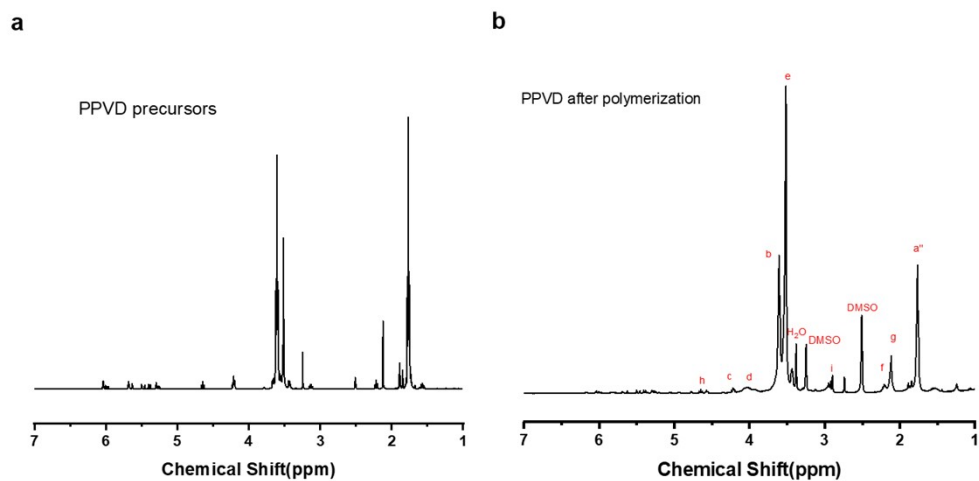


Figure S6. ^1H NMR spectrum of (a) PPVD precursor and (b) polymerized PPVD.

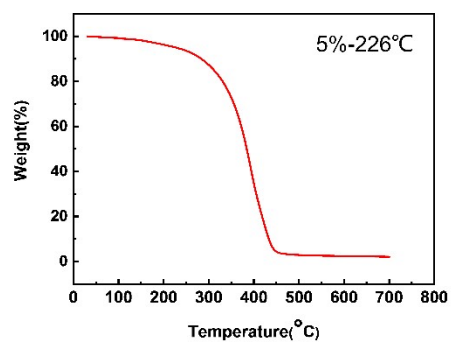


Figure S7. The thermogravimetric analysis of PPVD.

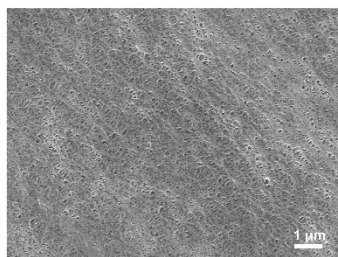


Figure S8. SEM image of 4 μm ultra-thin film.

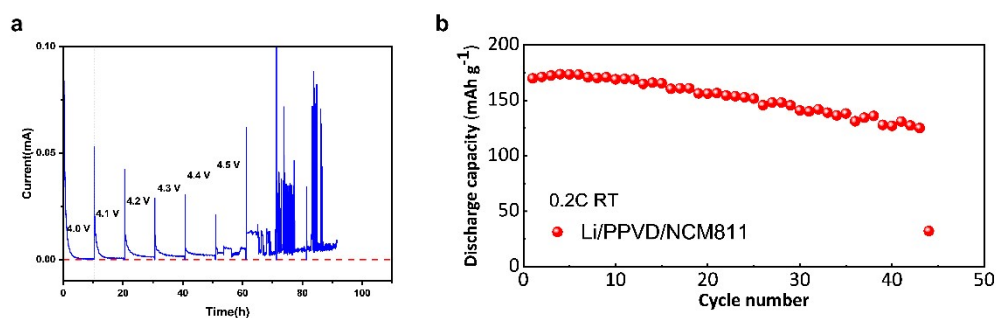


Figure S9. (a) Electrochemical floating analysis of the PPVD; (b) Cycling performance of the Li/PPVD/NCM full cell at 0.2 C

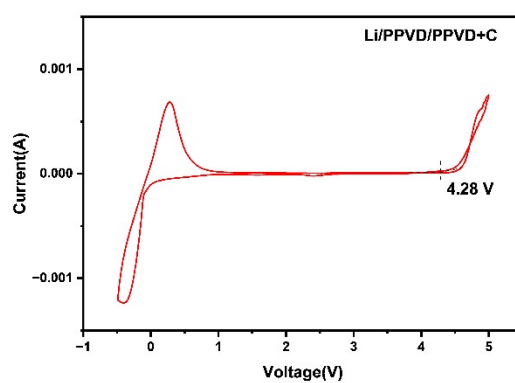


Figure S10. CV curve of Li/PPVD/PPVD-C cell within the voltage range of 2.5–5.0 V.

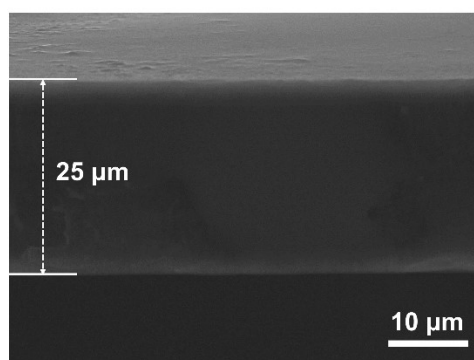


Figure S11. Cross-sectional SEM image of PPVD@PPF electrolyte.

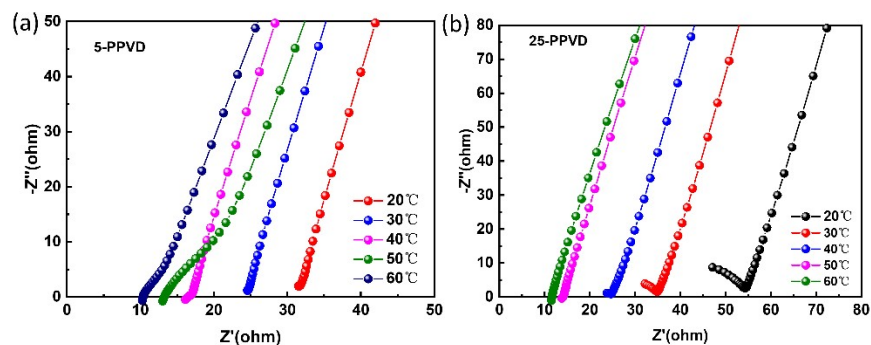


Figure S12. The EIS curves of symmetric cells with (a) PPVD@PEF and (b) PPVD@PPF as electrolyte.

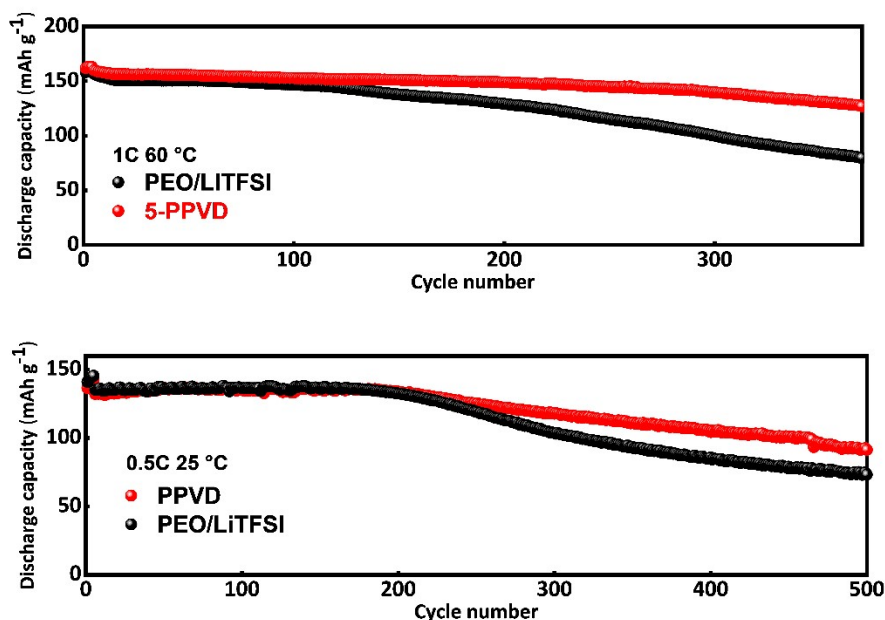


Figure S13. Cycling performance of Li|PPVD@PEF|LFP and Li|PEO@PEF|LFP cells at the current density of (a) 1 C and (b) 0.5 C.

Table S1. Corresponding ionic conductivity and conductance of polymer electrolytes with different molar ratio of EO:Li⁺

Molar ratio	6:1	4:1	2:1	1:1	1:2
Ionic conductivity(mS/cm)	0.0143	0.0188	0.0244	0.0206	0.103
Ionic conductance(S)	0.0676	0.0709	0.0766	0.0973	0.278

Table S2. The electrolytes with different molar ratios of PEGMEMA:VEC:DMAPMA

Electrolyte	PEGMEMA:VEC:DMAPMA
1	0.5:1:1
2	2:1:1
3	1:0.5:1
4	1:2:1
5	1:1:0.5
6	1:1:2
7	1:1:1

Table S3. Electrochemical performance of Li||LiFePO₄ cells at current density of 0.5 C with different ratio of monomers

Electrolyte	Initial capacity (mAh g ⁻¹)	Capacity retention @ cycles
1	146.3	75.3% @179 cycles
2	148.4	90.1% @152 cycles
3	149.5	91 % @219 cycles
4	151.2	93.2 % @210 cycles
5	159.5	79% @229 cycles
6	155.8	20.8% @50 cycles

Table S4. Ionic conductivity(σ) and Ionic conductance(G) for PPVD@PEF and PPVD@PPF

	Thickness(cm)	R(Ω)	Area(cm ⁻²)	σ (mS cm ⁻¹)	G (mS)
PPVD@ PEF	0.0004	10.28	1.886	0.0206	97.276
	0.0007	9.63	1.886	0.0386	103.87
	0.0008	8.55	1.886	0.0496	116.95
	0.0005	10.08	1.886	0.0263	99.26
	0.0005	8.46	1.886	0.0313	118.15
	Average			0.03329	107.099
	Standard deviation			0.01125	9.84
PPVD@ PPF	0.0025	14.54	1.886	0.0912	68.78
	0.0033	11.46	1.886	0.1526	87.24
	0.0035	25.14	1.886	0.0738	39.78

	0.0035	18.01	1.886	0.1030	55.52
	0.0033	15.98	1.886	0.1095	62.58
Average				0.10603	62.777
Standard deviatio n				0.02938	17.43