Supporting Information

A Borate Concentrated, Crosslinked Gel Polymer Electrolyte with Near-Single Ion Conduction for Lithium Metal Batteries

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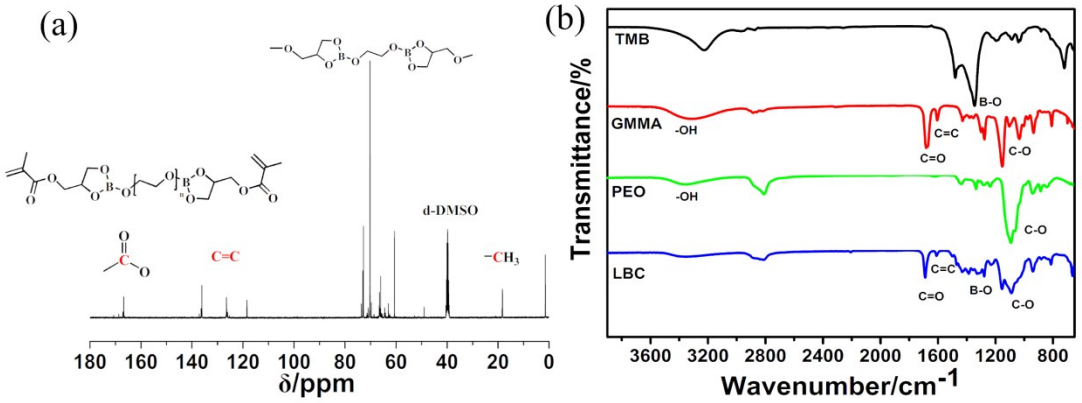

Figure S1. (a) ^{13}C-NMR spectrum of the LBC (b) FT-IR spectra of the TMB, PEG, GMMA and LBC.

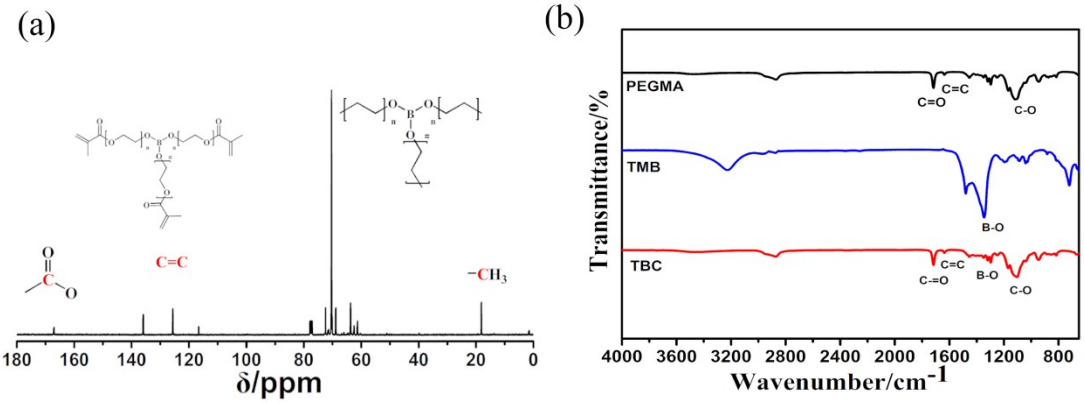

Figure S2. (a)^{13}C-NMR spectrum of TBC (b) FT-IR spectra of TMB, PEGMA and TBC.

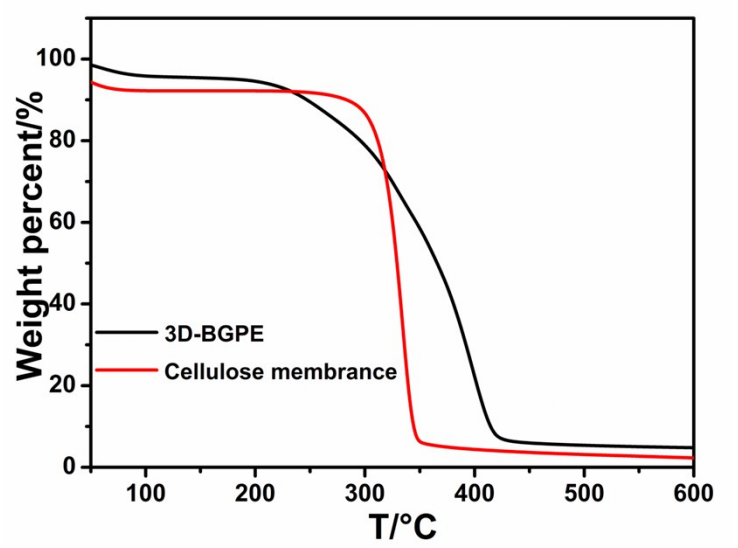

Figure S3. TGA thermograms of 3D-BGPE and cellulose membrane.

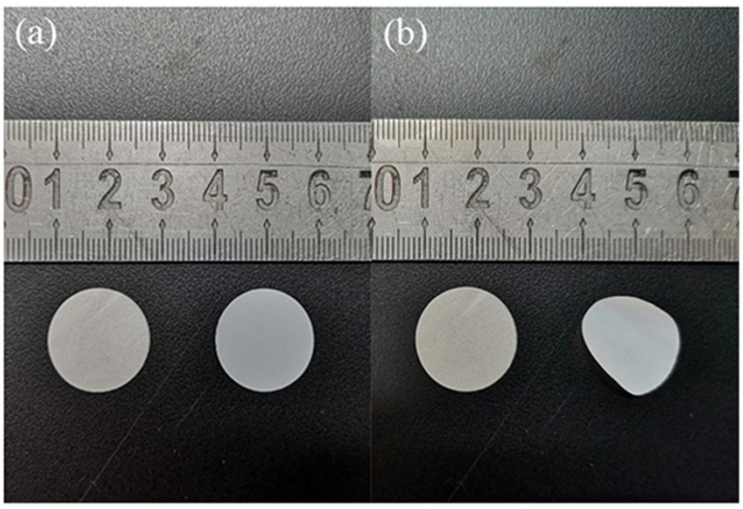

Figure S4. Thermal shrinkage of the 3D cross-linking membrane (left) and a commercial separator (right) (a) before and (b) after exposure to 150°C for 10 min.

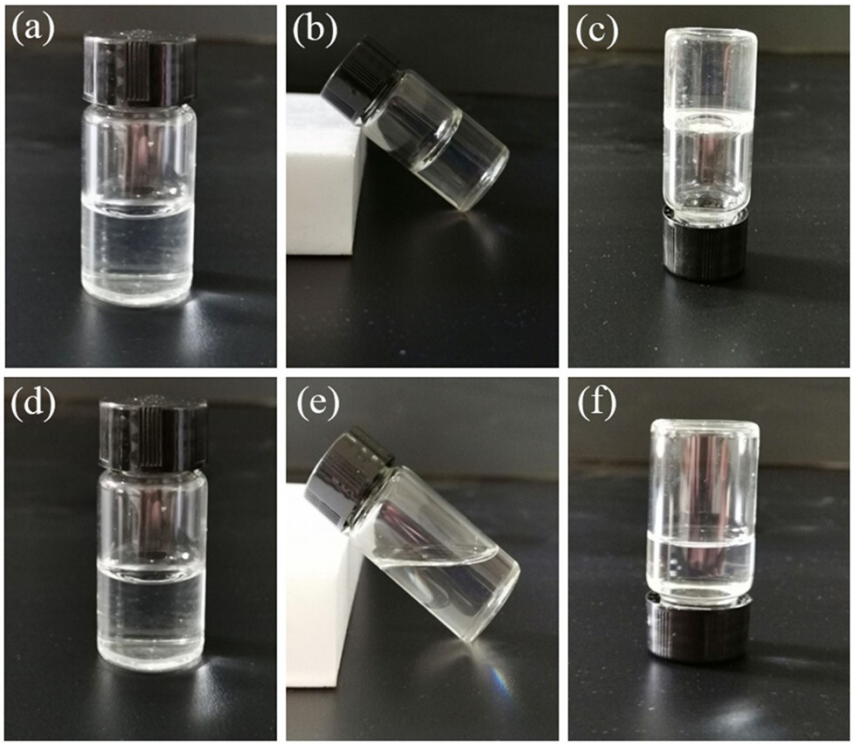

Figure S5. Optical photographs of the LE and 3D-BGPE.

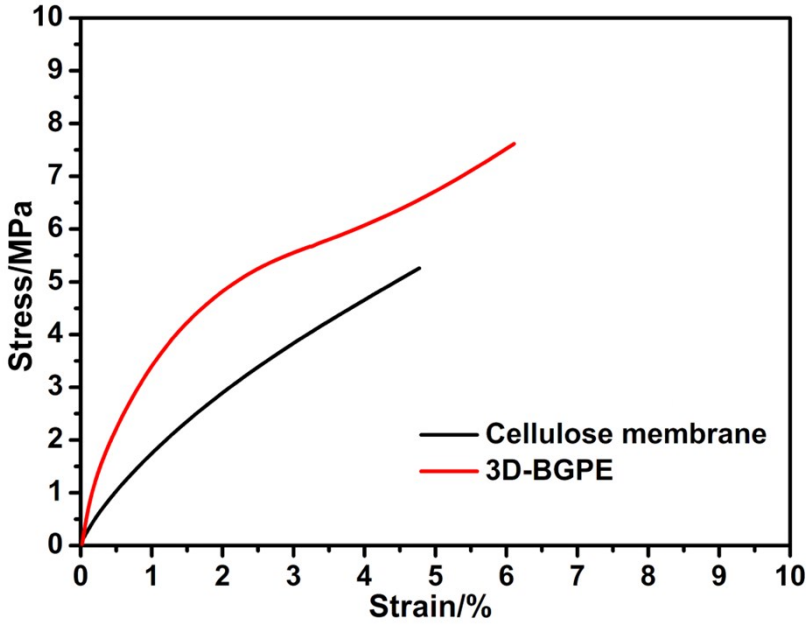

Figure S6. Stress-strain curves of 3D-BGPE and cellulose membrane.

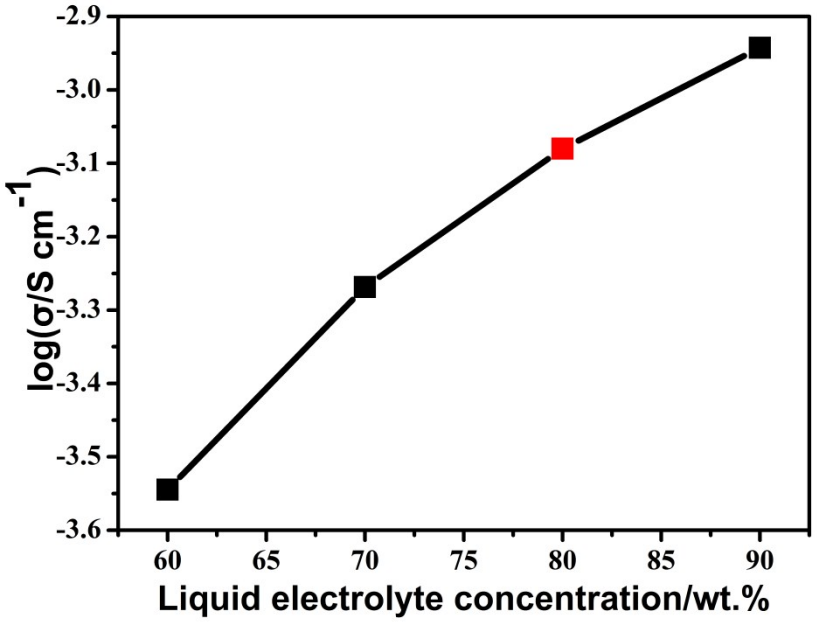

Figure S7. Ionic conductivity dependence on EC/DMC concentration for gel polymer electrolyte at 30 °C.

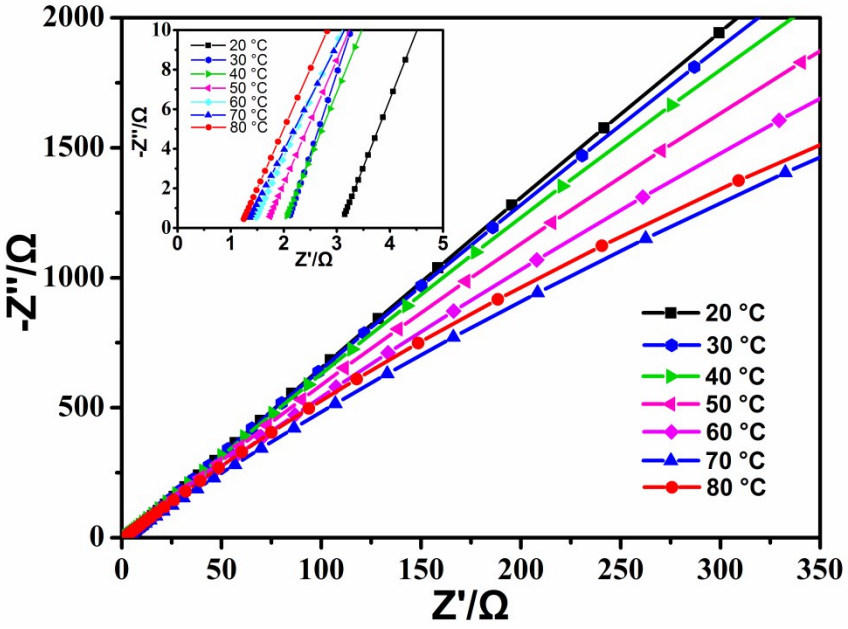

Figure S8. EIS of a SS|3D-BGPE|SS symmetrical cell with the elevation of temperature.

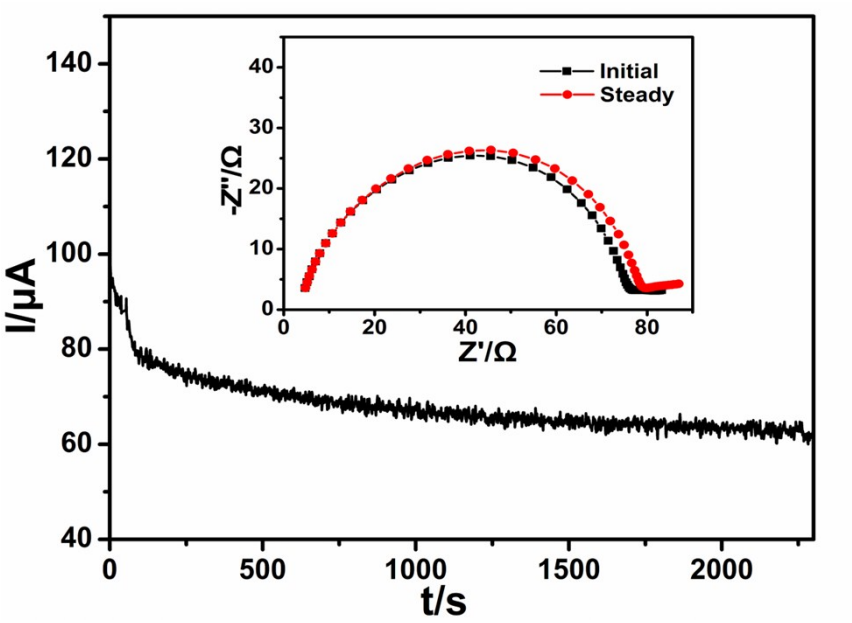

Figure S9. Current variation with polarization of a Li|LE|Li symmetrical cell with an applied potential of 10 mV and EIS before and after polarization.

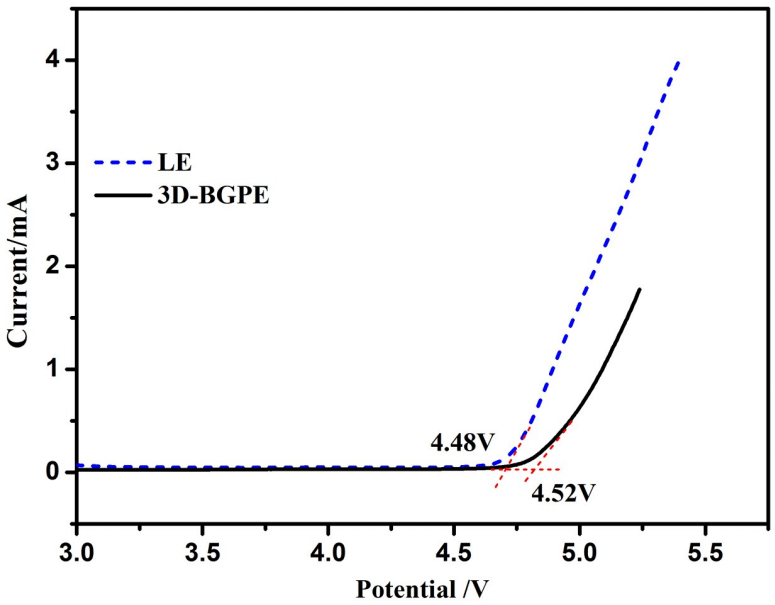

Figure S10. Linear sweeping voltammetry curves of the Li/3D-BGPE/SS cell and Li/LE/SS cell at 30 °C.

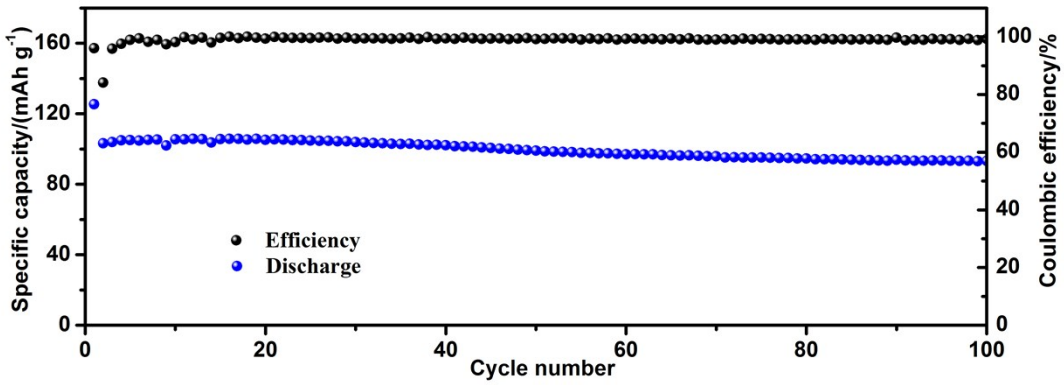

Figure S11. Cycling performance of $LiFePO_4$/3D-BGPE/ Li cell at 2C at 30 °C.

Table S1. EIS results of the LiFePO4 /3D-BGPE/ Li and LiFePO4/LE/Li cells after 10 cycles and 50 cycles at 0.5 C

Battery sample	After 10 cycles			After 50 cycles		
	R_b	R_f	R_{ct}	R_b	R_f	R_{ct}
3D-BGPE	2.95	39.0	41.0	3.65	41.7	42.1
Liquid electrolyte	2.75	10.5	23.1	4.62	26.5	32.9

The EIS curves were analyzed using an equivalent circuit by Z-view software. In the equivalent circuit, R_b represents bulk resistance, R_f represents interface resistance, R_{ct} represents charge transfer resistance, Z_w represents Warburg impedance, and CPE1 and CPE2 are the constant phase elements. The simulation results are summarized in **Table S1**. It is apparently seen that the $LiFePO_4$/3D-BGPE/Li cell displays an interfacial resistance (R_f and R_{ct}) with negligible variance after 10 cycles and 50 cycles, while the interfacial resistance of $LiFePO_4$/LE/Li cell increases noticeably. The negligible variance in interfacial resistance suggests that the interface between electrode and 3D-BGPE keeps steady during charge-discharge cycles, which leads to the superior cycling stability of $LiFePO_4$/3D-BGPE/Li cell as shown in Figure 6c.