

***In-situ* Forming Asymmetric Bi-functional Gel Polymer Electrolyte in Lithium-Sulfur Batteries**

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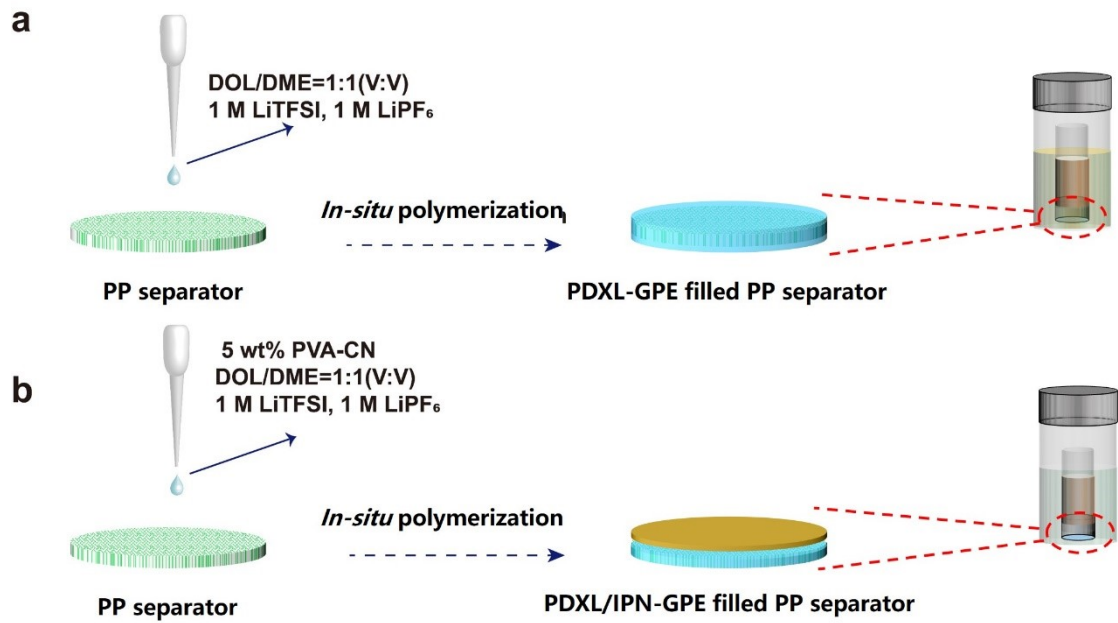


Fig. S1 The schematic diagrams of preparing PDXL-GPE and PDXL/IPN-GPE filled PP separator for UV-vis spectra.

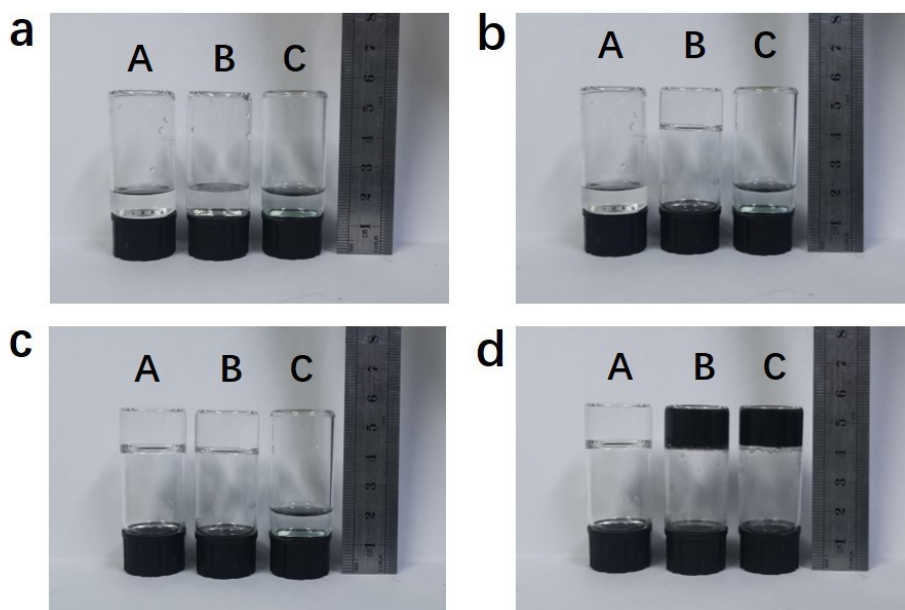


Fig. S2 The photos of gelation process. Precursor solutions after (a) 0 hour, (b) 12 hours, (c) 24 hours at room temperature and (d) after 6 hours at 70 °C. *A*: 1 M LiPF₆ and 1.0 M LiTFSI in DOL/DME (1:1, v/v), *B*: *A*+5 wt% PVA-CN, *C*: *B* without DOL.

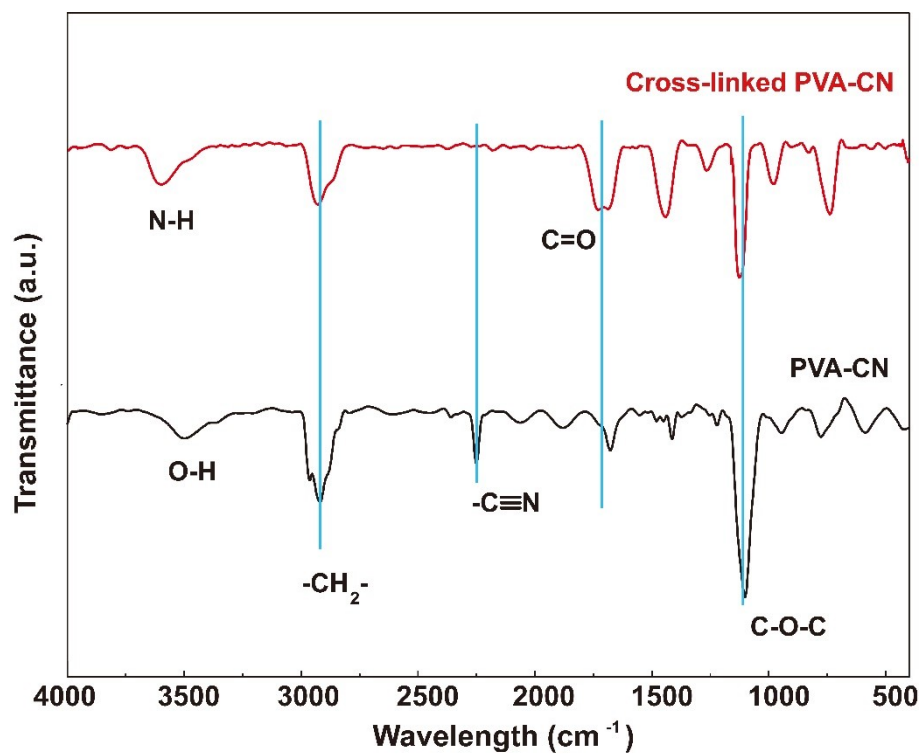


Fig. S3 The FTIR spectra of PVA-CN (black line) and cross-linked PVA-CN (red line).

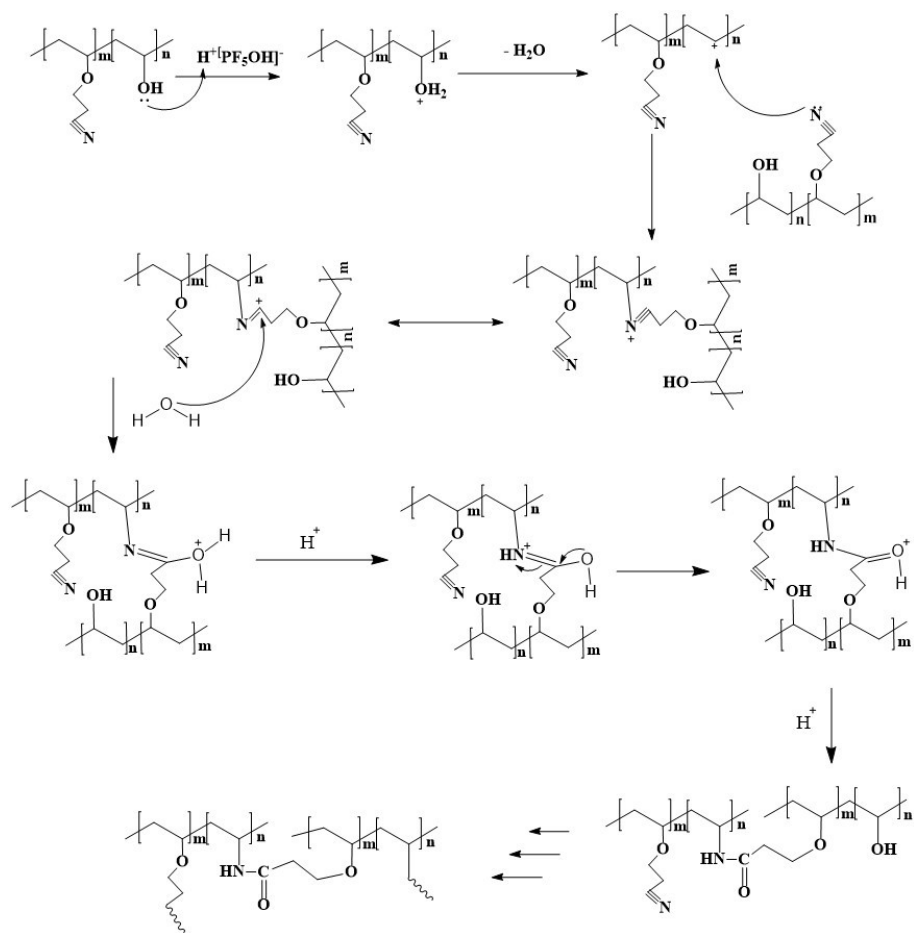


Fig. S4 Possible reaction procedures following Ritter reaction mechanism.

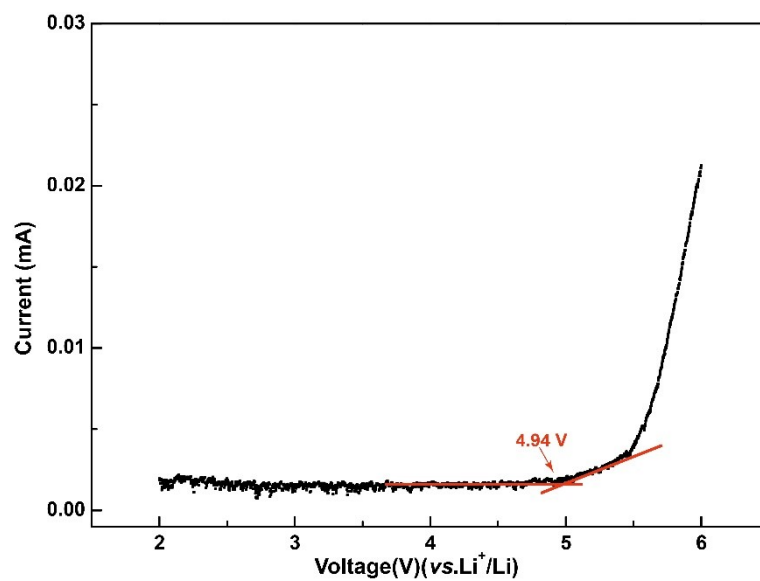


Fig. S5 The LSV of IPN-GPE at a scanning rate of 0.1 mV s^{-1} .

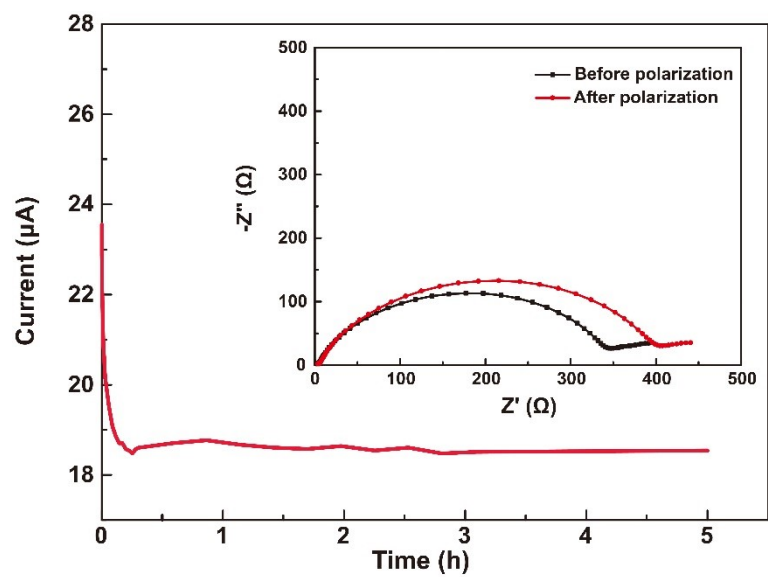


Fig. S6 The chronoamperometry profiles of the symmetric Li|PDXL-GPE|Li cell under a polarization voltage of 10 mV, and the EISs before and after polarization (inset).

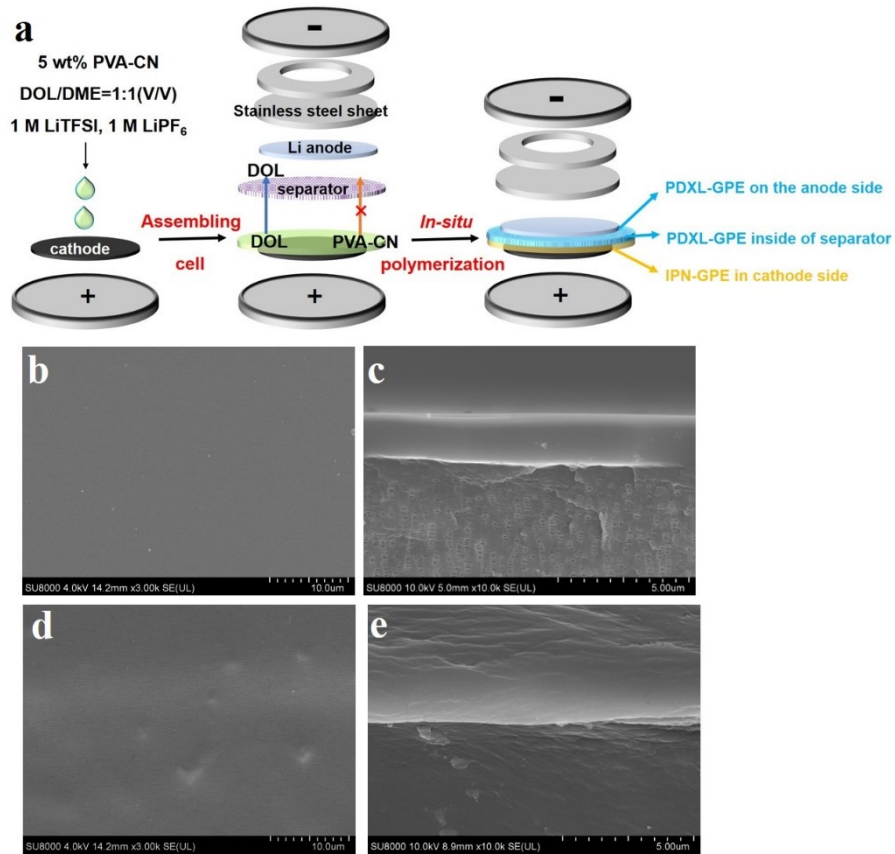


Fig. S7 (a) The schematic illustration of Li-S cells with the asymmetric GPE. (b) Surface and (c) cross-section of cross-linked PVA-CN-GPE on the surface of PP separator with the gelation of LE(C). (d) Surface and (e) cross-section of PDXL/IPN-GPE filled PP separator with the gelation of LE(B).

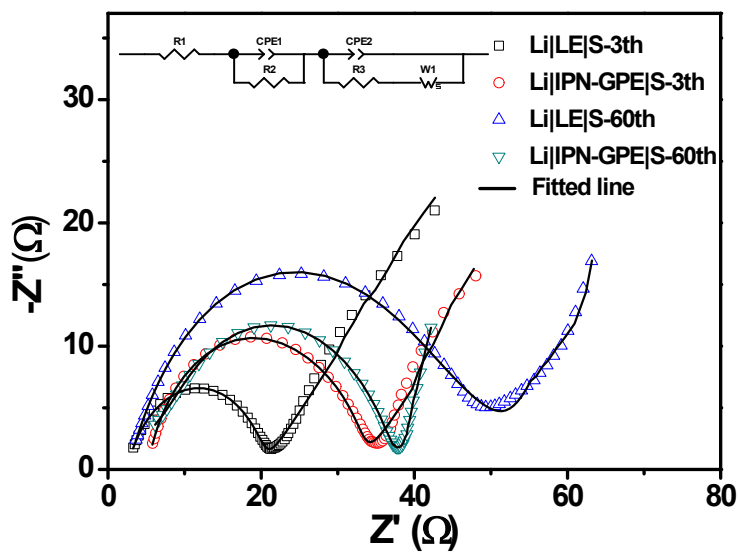


Fig. S8 Nyquist profiles and fitted EIS plots of the Li|LE|S and Li|IPN-GPE|S batteries after the first 3 cycle's activation at 0.1 C and 60 cycles at 0.5 C.

Table S1 EIS fitted results of Li-S batteries

Cycle number	Li LE S			Li IPN-GPE S		
	R_b, Ω	R_{SEI}, Ω	R_{ct}, Ω	R_b, Ω	R_{SEI}, Ω	R_{ct}, Ω
3th	2.3	8.0	9.8	5.1	1.3	25.2
60th	2.3	18.9	33.4	3.5	2.9	30.8

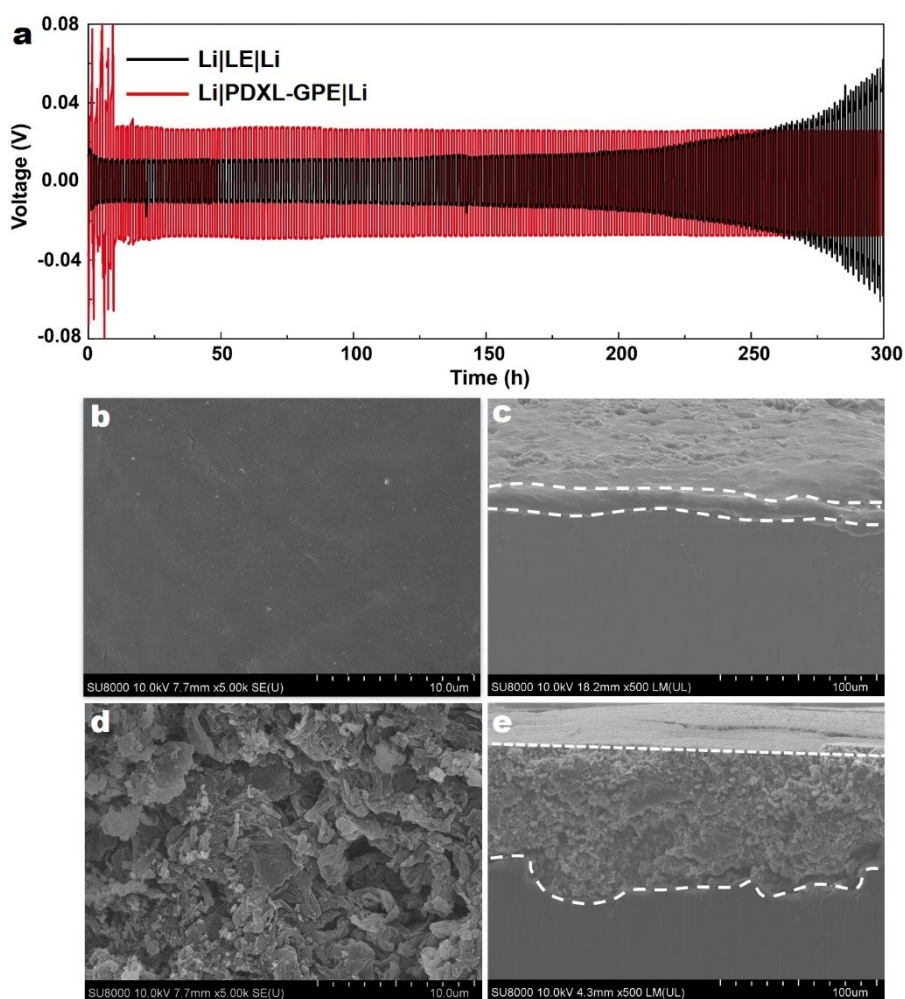


Fig. S9 (a) Voltage profiles of Li||Li symmetric cells with LE and PDXL-GPE at 1.0 mA cm^{-2} and 1.0 mAh cm^{-2} . (b) Surface and (c) cross-sectional SEM images of Li metal anodes in Li|PDXL-GPE|Li cells after 150 cycles. (d) Surface and (e) cross-sectional SEM images of Li metal anodes in Li|LE|Li cells after 150 cycles.

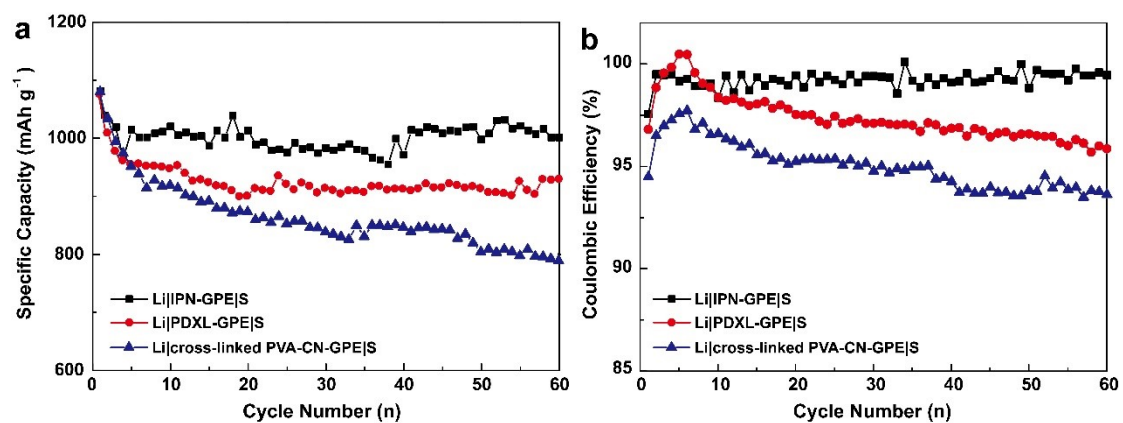


Fig. S10 (a) Cycling performances and (b) CEs of Li|IPN-GPE|S, Li|PDXL-GPE|S and Li|cross-linked PVA-CN-GPE|S batteries at 0.1 C for the first three cycles and 0.5 C for the later cycles.