

Supplementary Information

A Hybrid Solid-State Electrolyte Endows Li Metal Battery with Excellent Cycling Life at 120 °C

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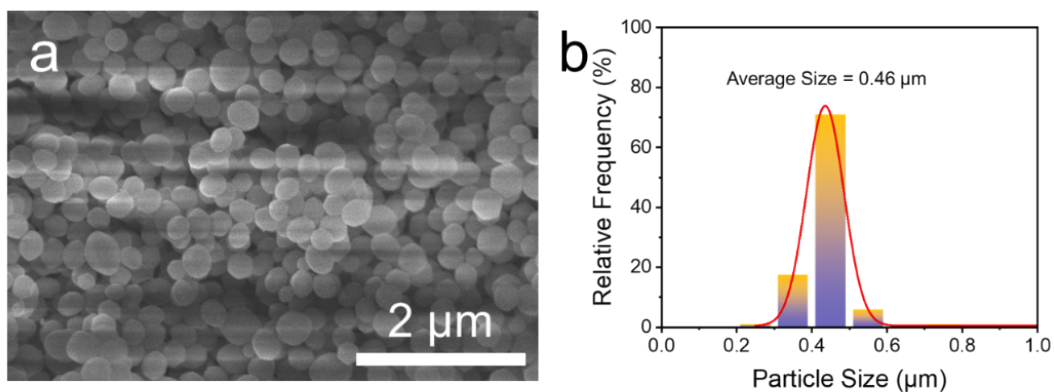


Fig. S1 (a) SEM image and (b) particle size distribution of MOF powder.

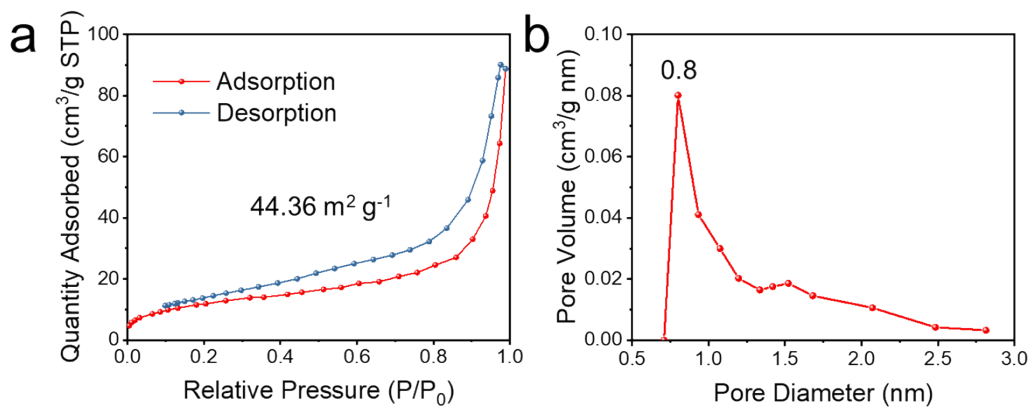


Fig. S2 (a) Nitrogen adsorption/desorption isotherms and (b) pore size distribution of MOF.

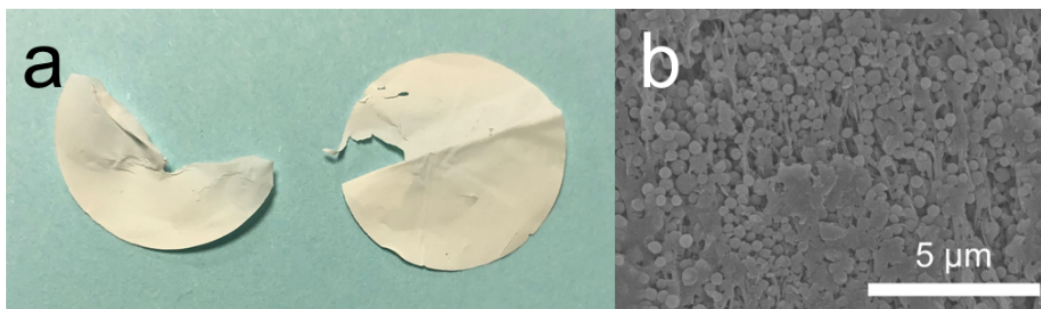


Fig. S3 (a) MOF film with a ratio of 4:1 was poorly tough and easy to break; (b) MOF film with a ratio of 2:1 had too much PVDF to cover the MOF.

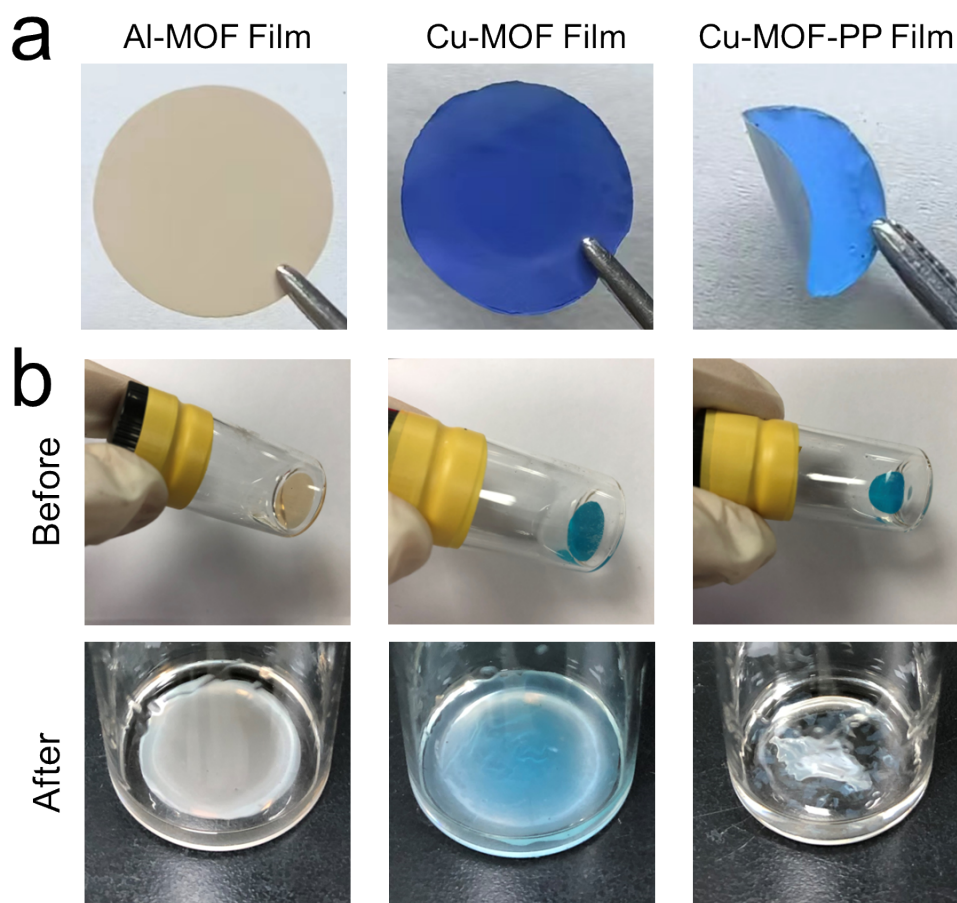


Fig. S4 (a) Photographs of Al-MOF film, Cu-MOF film, and Cu-MOF-PP film; (b) Photographs of the three films immersed in electrolyte before and after heating at 120 °C.

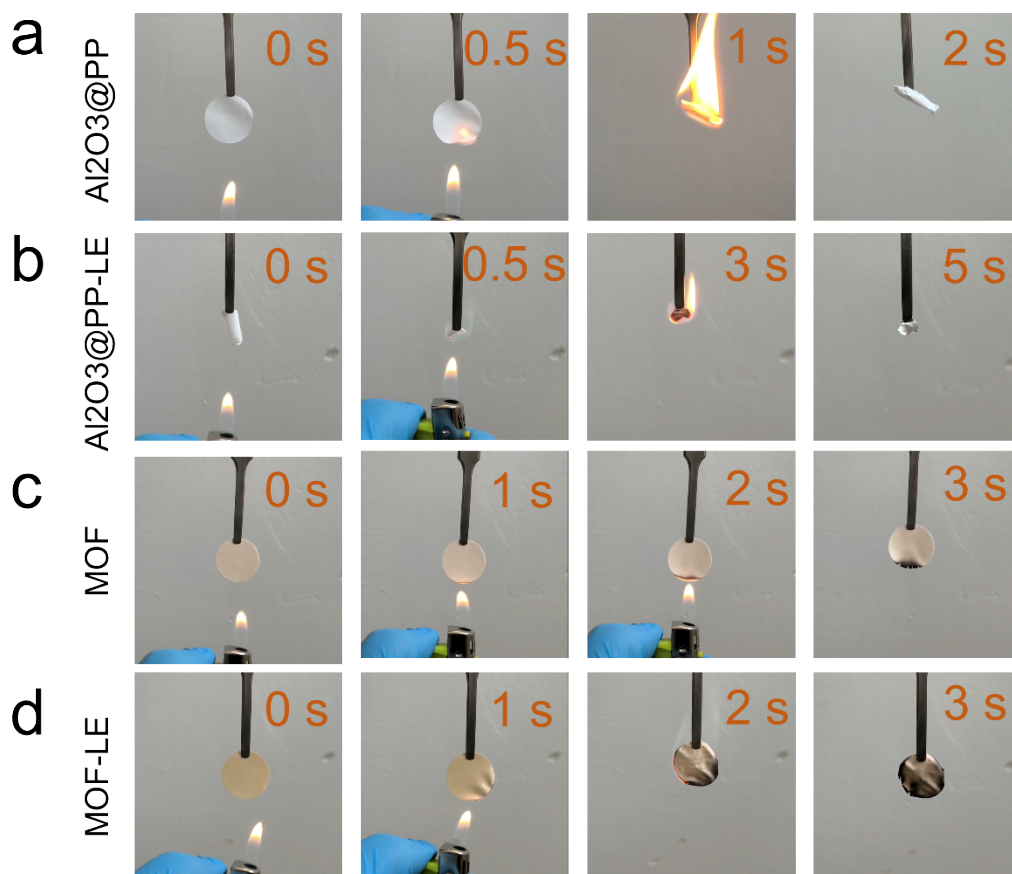


Fig. S5 Ignition tests of $\text{Al}_2\text{O}_3@PP$ separator and MOF film before and after infiltration of electrolyte.

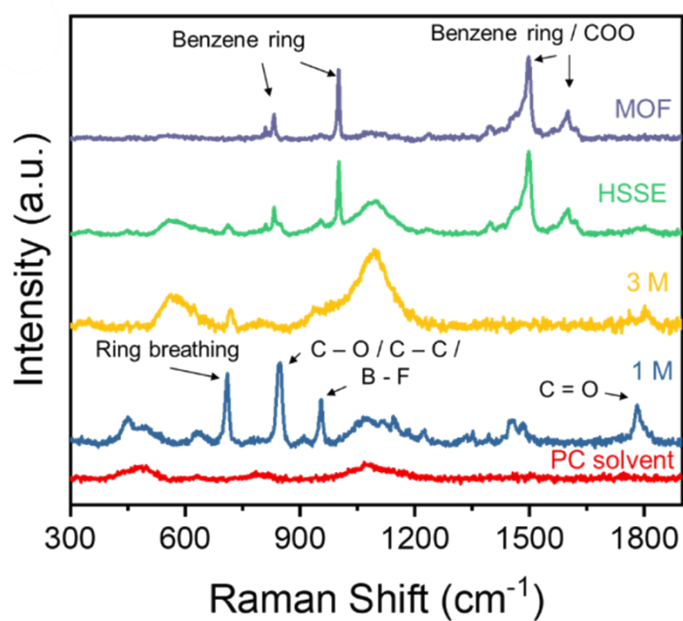


Fig. S6 Raman spectra of PC solvent, 1 M and 3 M LiDFOB-PC, MOF, and HSSE in the range of 300 to 1900 cm^{-1} .

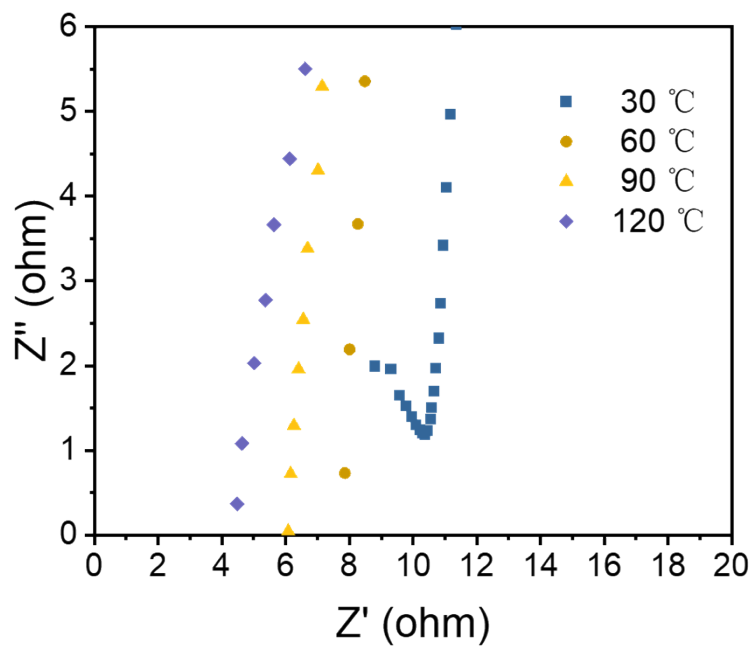


Fig. S7 Nyquist plots of SS|HSSE|SS at different temperatures.

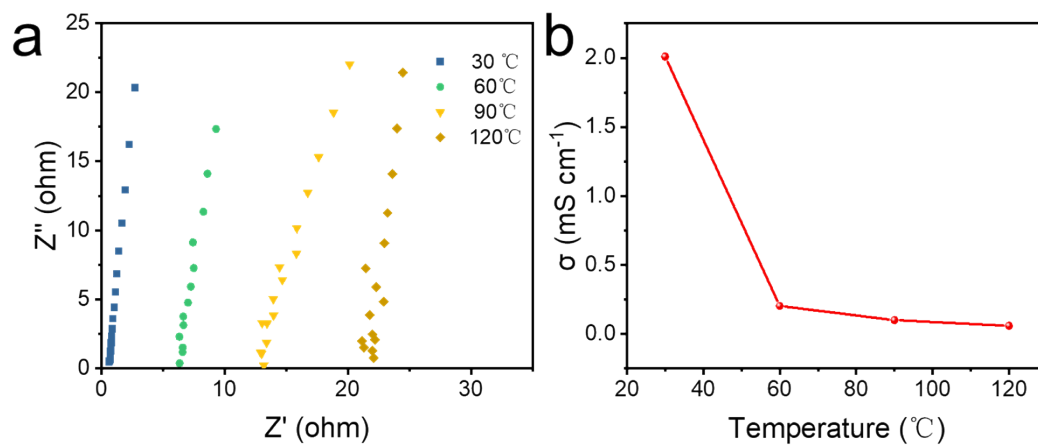


Fig. S8 (a) Nyquist plots and (b) ionic conductivity of PP-LE at different temperatures.

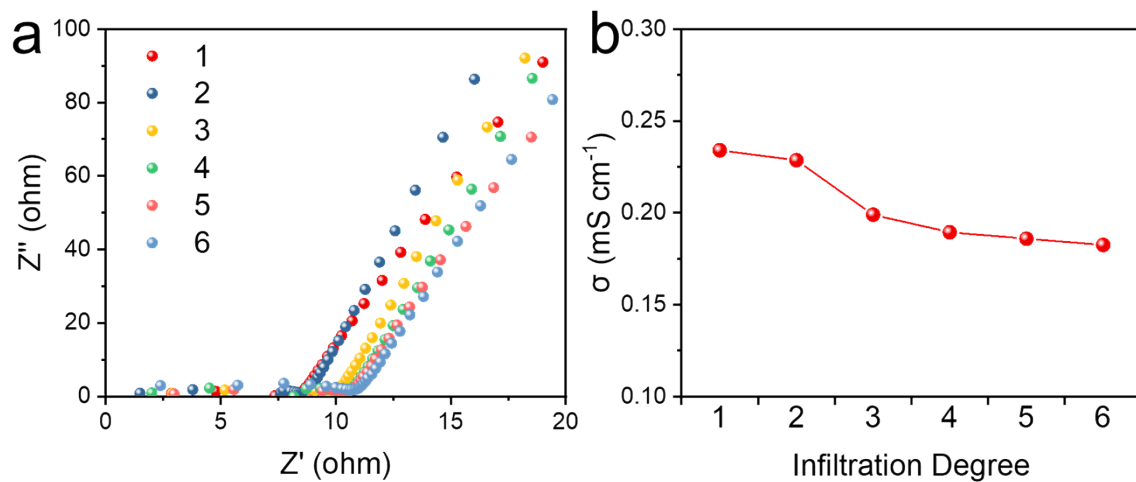


Fig. S9 EIS and ionic conductivity measured at different infiltration degrees.

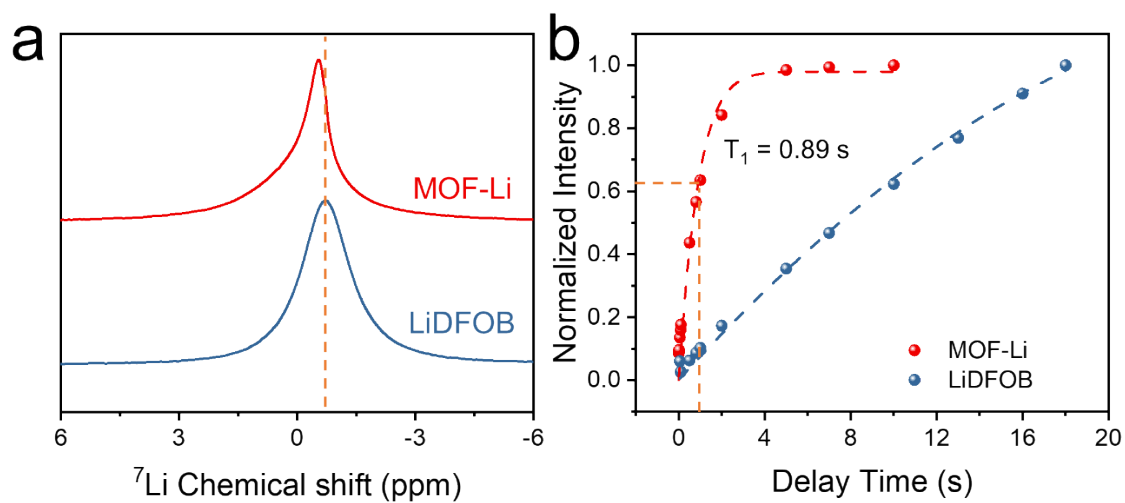


Fig. S10 (a) ⁷Li ssNMR spectra and (b) the fitting result of T_1 relaxation time of MOF-Li and LiDFOB from ⁷Li ssNMR.

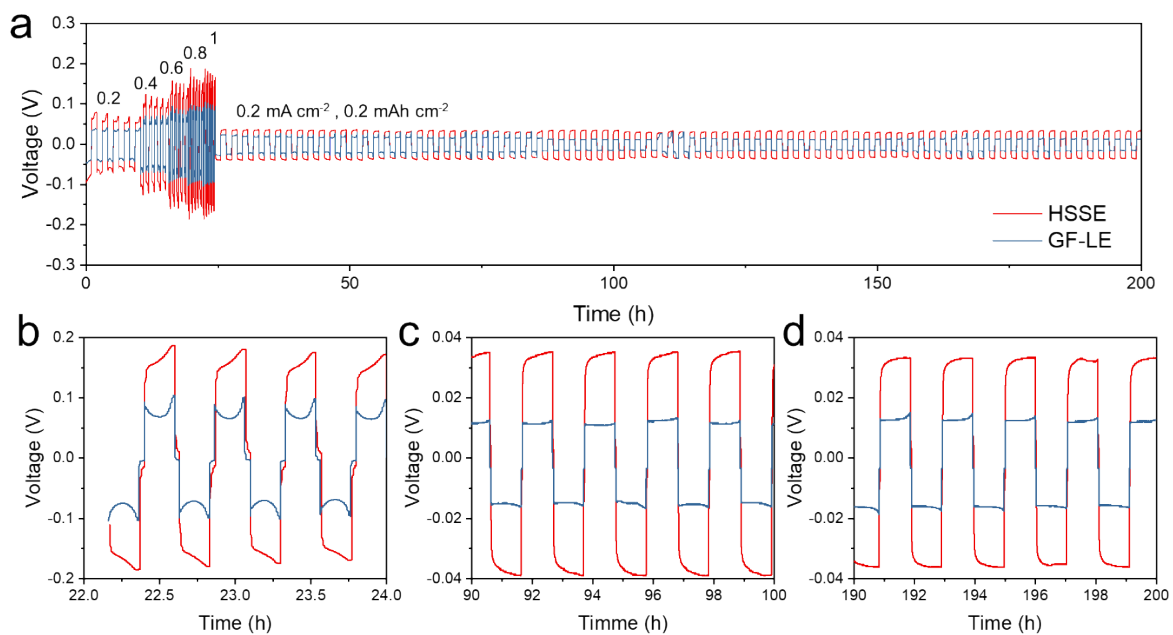


Fig. S11 Symmetrical cell performances and EIS of the Li|HSSE|Li and Li|GF-LE|Li symmetric cells.

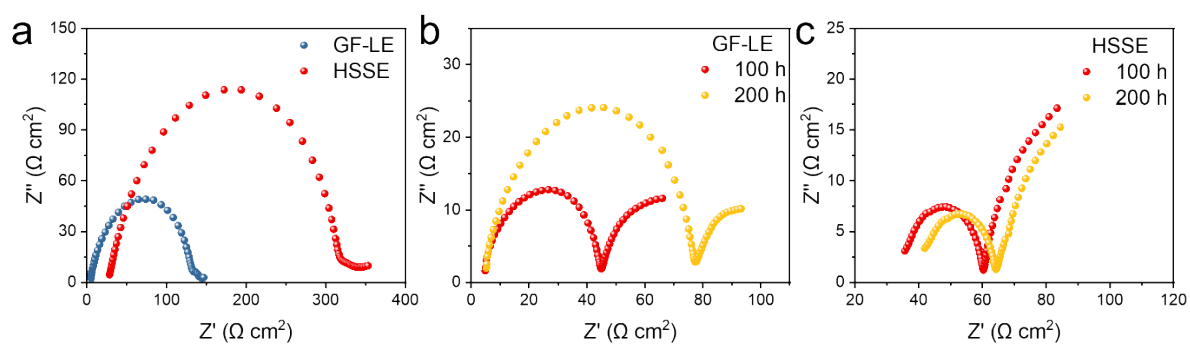


Fig. S12 The initial and after-cycle impedance of Li|HSSE|Li and Li|GF-LE|Li.

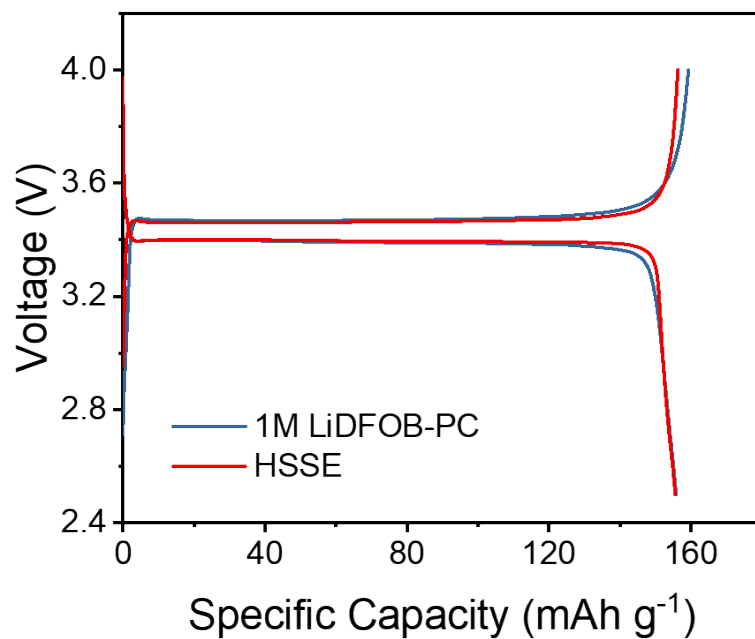


Fig. S13 First charge/discharge curves of LFP|GF-LE|Li and LFP|HSSE|Li at room temperature and 0.2 C current density.

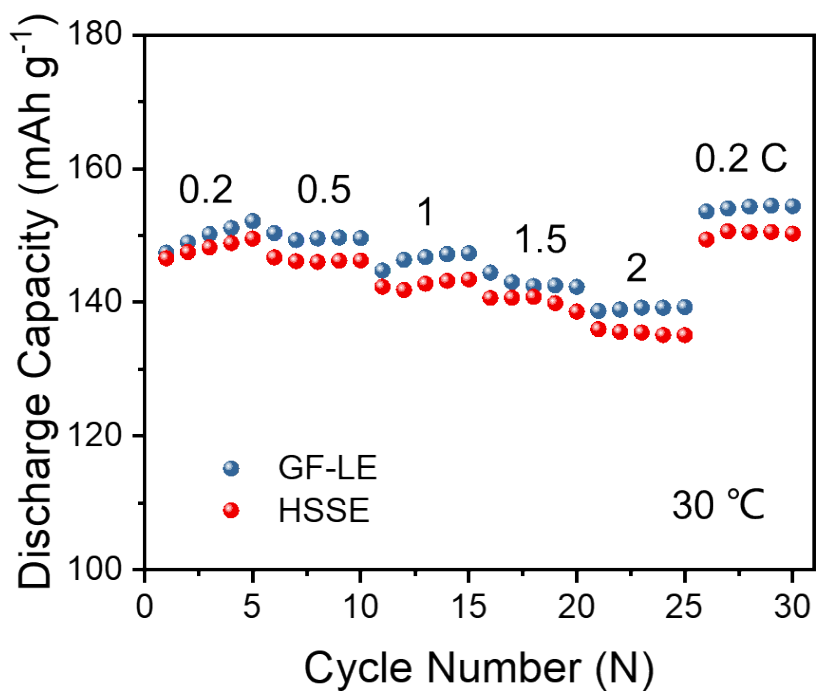


Fig. S14 Rate performance of LFP|HSSE|Li and LFP|GF-LE|Li cells at room temperature

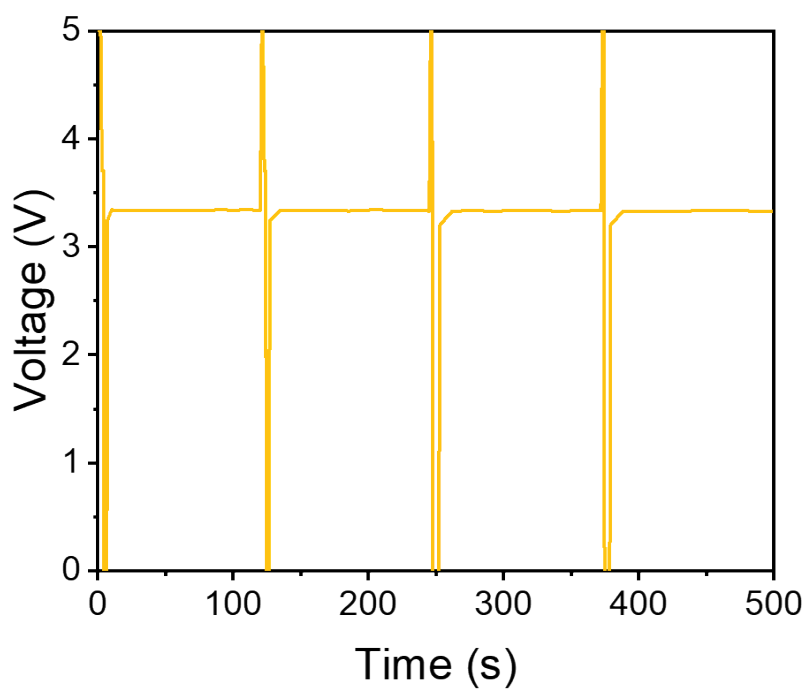


Fig. S15 Voltage profiles of LFP|HSSE(Cu)|Li cells at 120 °C.

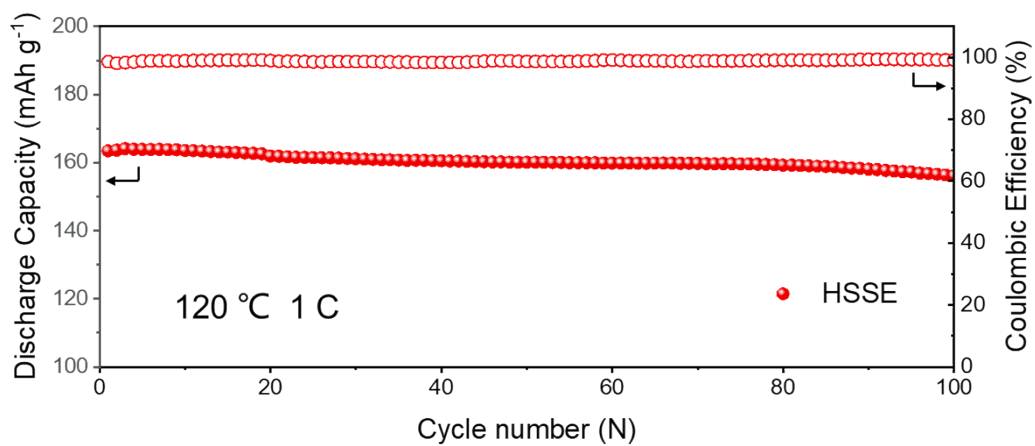


Fig. S16 Cycle performance of LFP|HSSE|Li battery at 120 °C and 1 C current density.

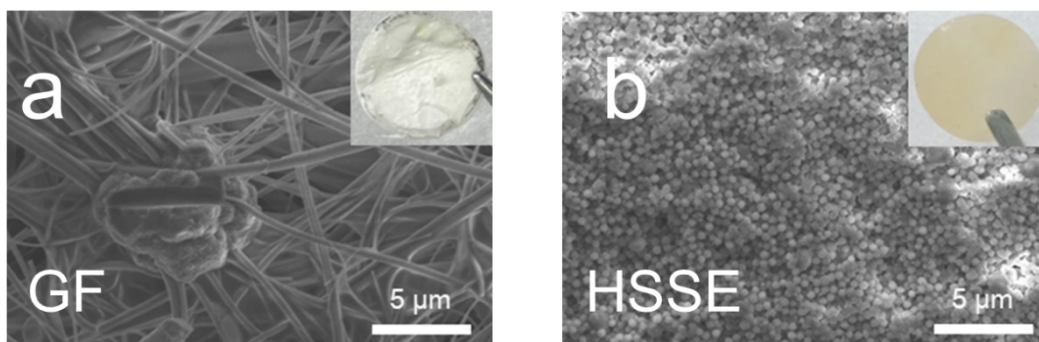


Fig. S17 Photographs and SEM images of peeled (a) glass fiber separator and (b) HSSE after cycling at 120 °C.

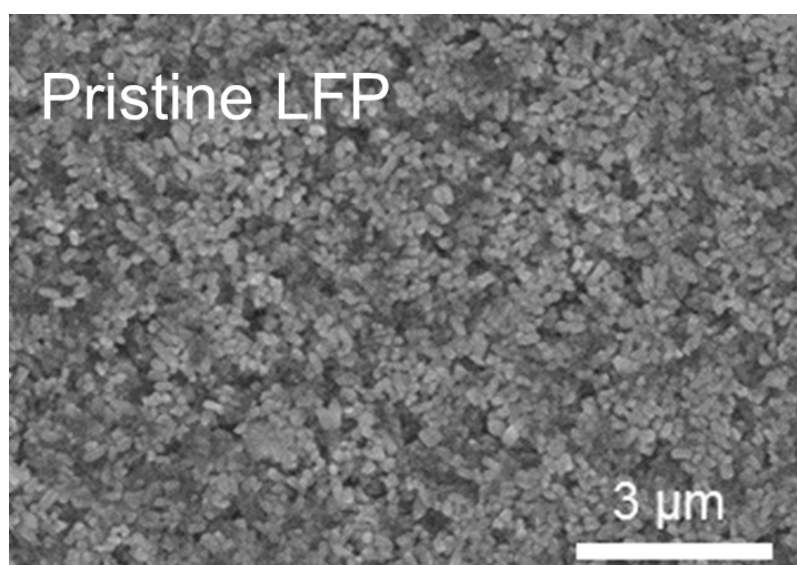


Fig. S18 SEM image of the pristine LFP cathode surface.

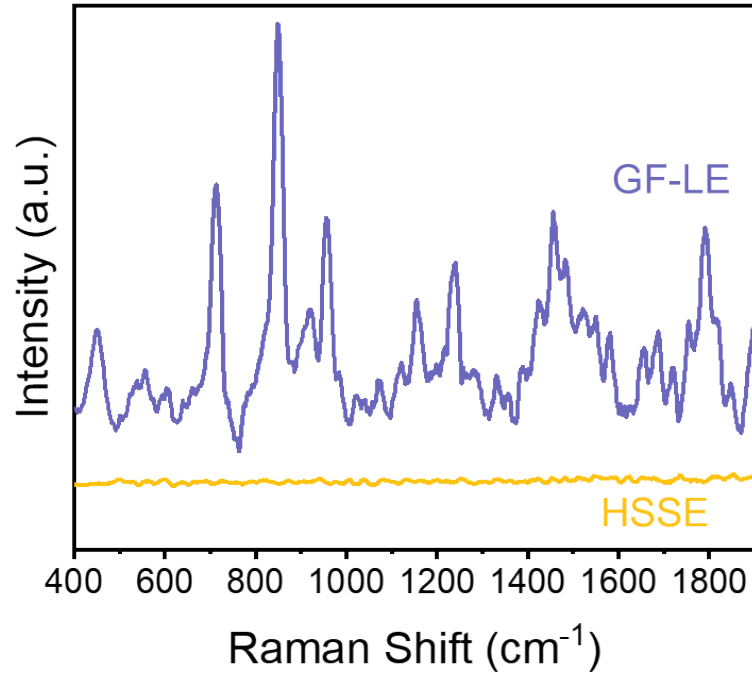


Fig. S19 Raman spectra of the surface of the lithium metal cathode after cycling in different electrolytes at 120 °C.

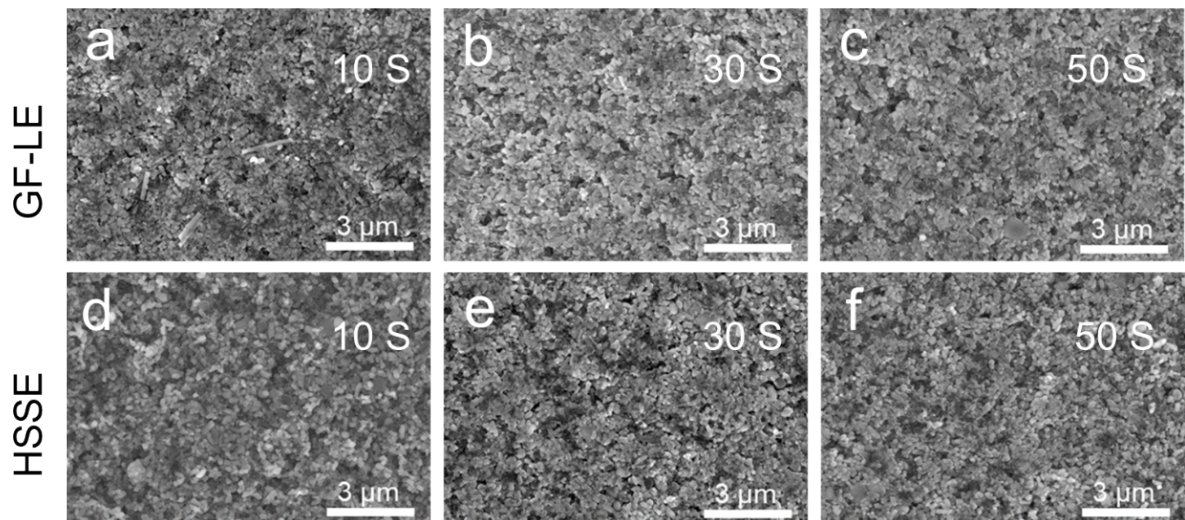


Fig. S20 SEM images of the argon etched LFP cathode surface after cycling in different electrolytes at 120 °C (a-c) GF-LE; (d-f) HSSE.

Table S1. Frontier molecular orbital theory analysis of PC and [Li-PC]⁺.

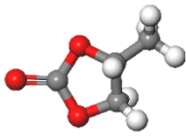
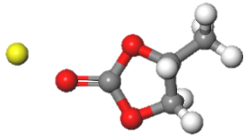




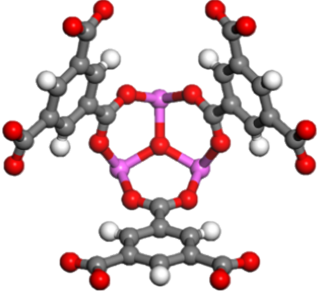
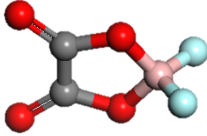
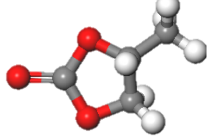
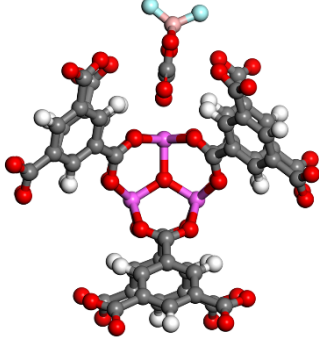
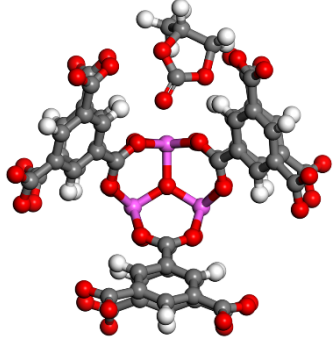






Electrolytes			
	PC	[Li-PC] ⁺	
LUMO (eV)	-0.60	-4.95	
HOMO (eV)	-8.36	-12.83	
 C	 O	 H	 Li ⁺

Table S2. DFT calculation of the adsorption energy of activated MIL-96(Al) clusters on DFOB⁻ and PC.

 Activated MIL-96(Al) clusters	 DFOB ⁻	 PC			
	E = -16365.517 eV	E = -10379.519 eV			
 MOF-DFOB ⁻	 MOF-PC				
		E = -168096.803 eV	E = -162105.464 eV		
E = -151718.872 eV	E = -168096.803 eV	E = -162105.464 eV			
E _{ad}	-12.412 eV	-7.071 eV			
 Al	 C	 O	 H	 B	 F