Hf-based UiO-66 type solid electrolytes for all-solid-state lithium batteries

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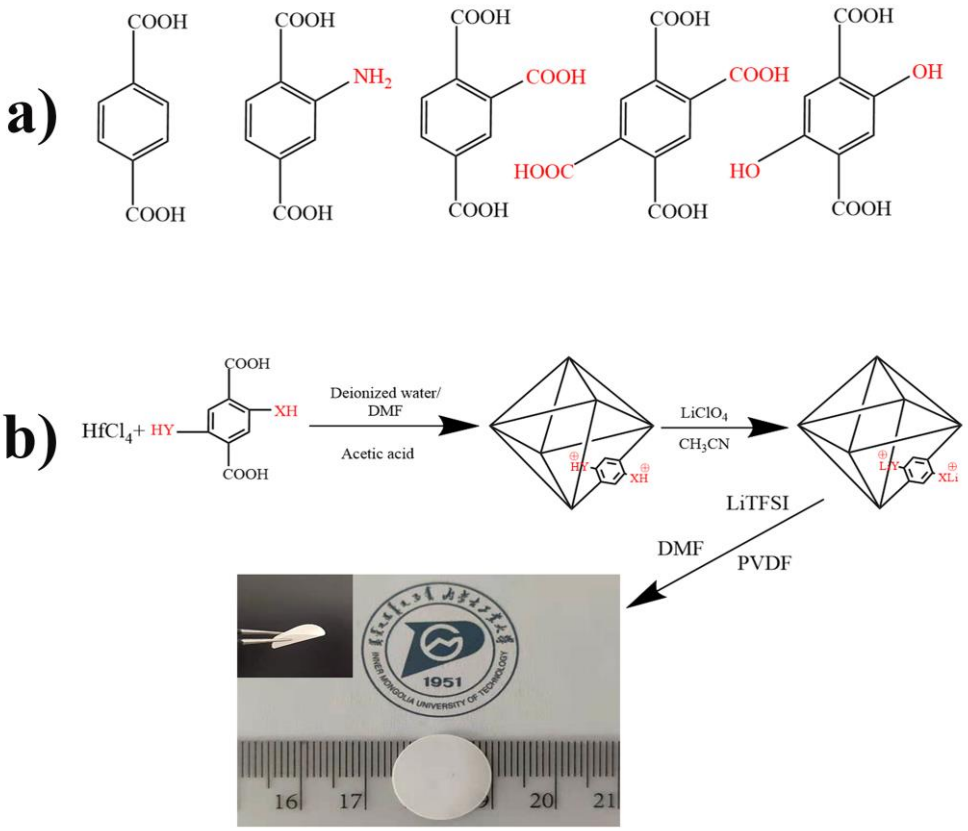

Fig. S1 a) Ligand of MOF-based SEs; b) Synthetic procedure of electrolyte membrane and photographs of Li/HLMOF-4 electrolyte membrane.

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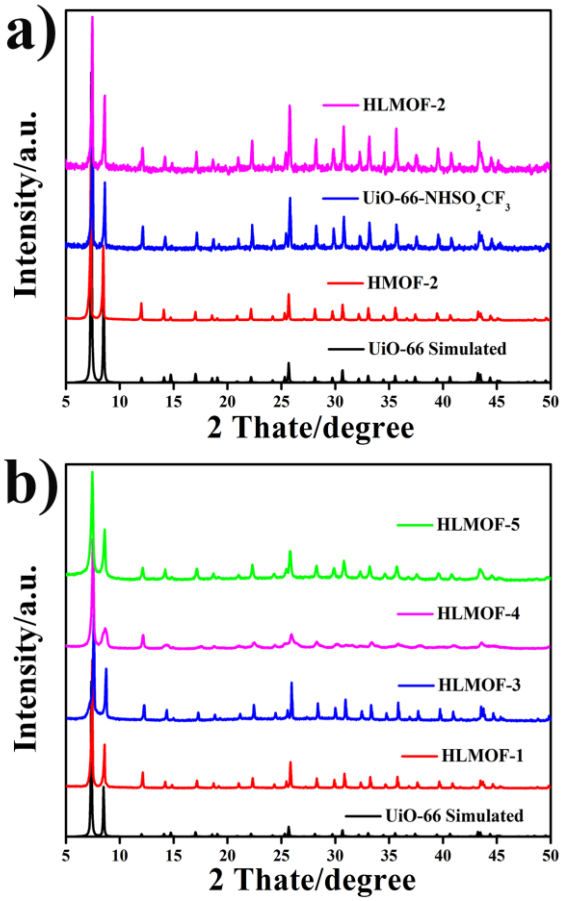

Fig. S2 The XRD of a) HLMOF-2; b) HLMOFs.

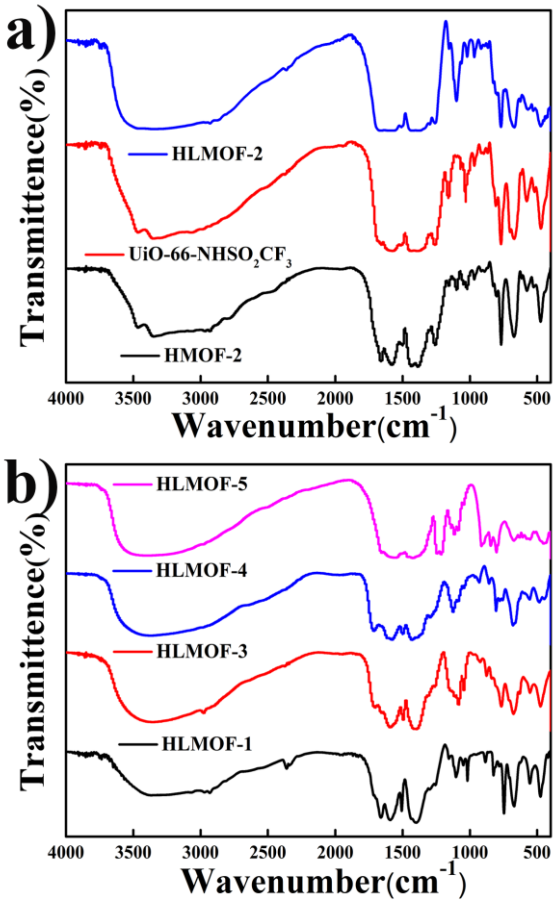

Fig. S3 FT-IR of a) HMOF-2, UiO-66-$NHSO_2CF_3$, HLMOF-2; b) HLMOFs.

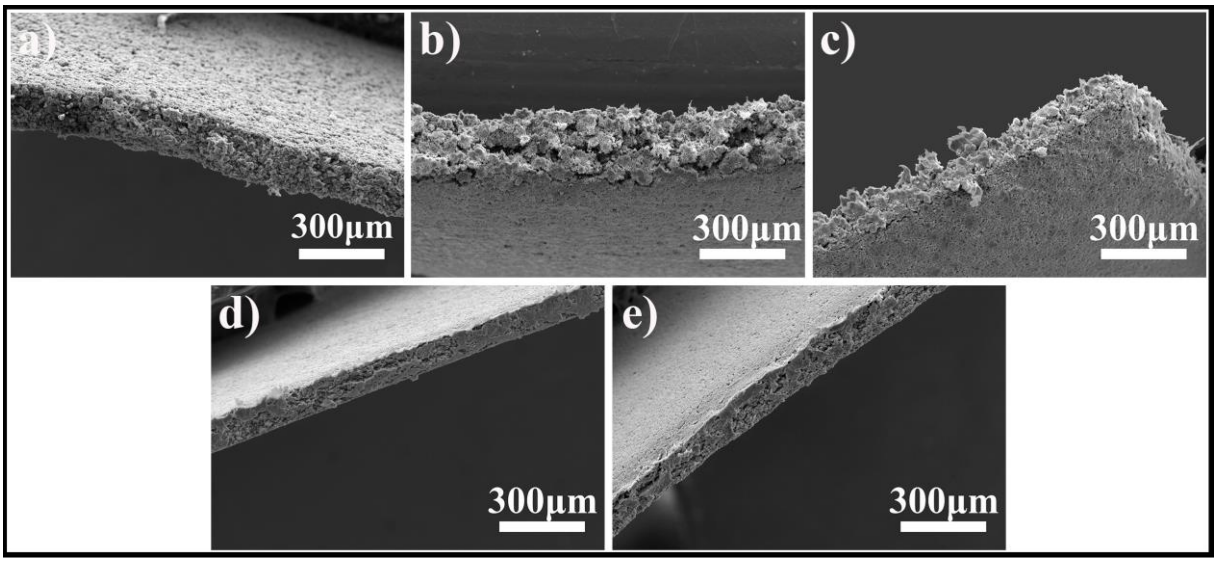

Fig. S4 The SEM of electrolyte membrane a) Li/HLMOF-1; b) Li/ HLMOF-2; c) Li/ HLMOF-3; d) Li/ HLMOF-4; e) Li/ HLMOF-5 at the scale of cross-sectional diagram.

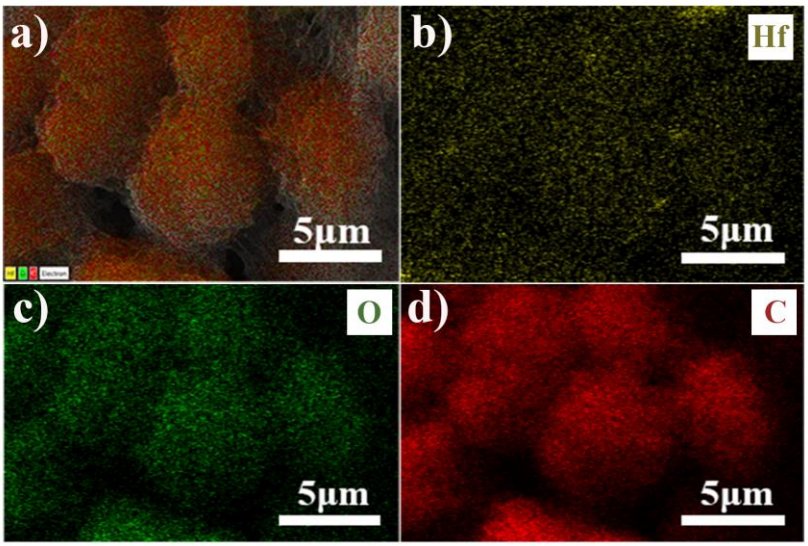

Fig. S5 EDS elemental analysis of the Li/HLMOF-1. a) EDS elemental mapping of all elements. EDS mapping of b) Hf element, c) O element and d) C element.

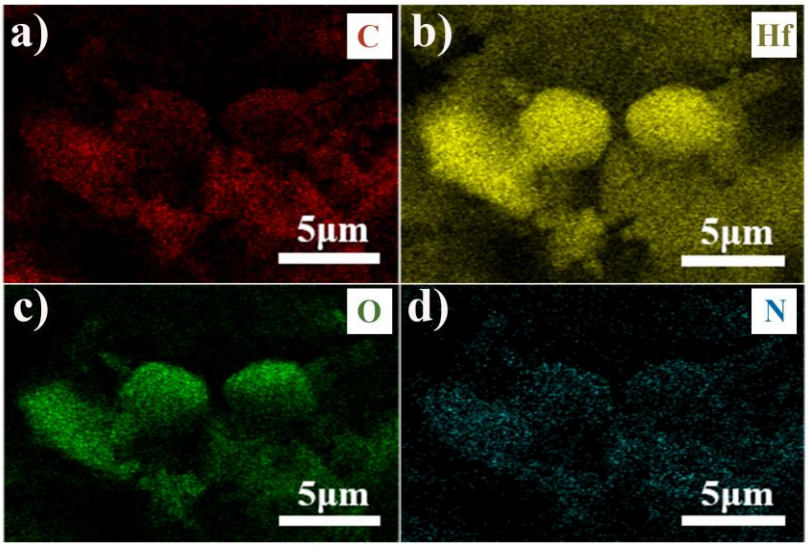

Fig. S6 EDS elemental analysis of the Li/HLMOF-2. EDS mapping of a) C element, b) Hf element, c) O element and d) N element.

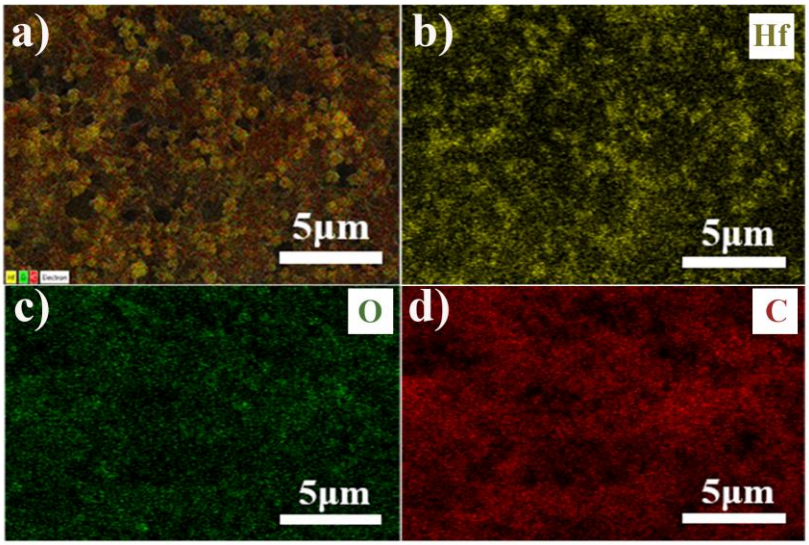

Fig. S7 EDS elemental analysis of the Li/HLMOF-3. a) EDS elemental mapping of all elements. EDS mapping of b) Hf element, c) O element and d) C element.

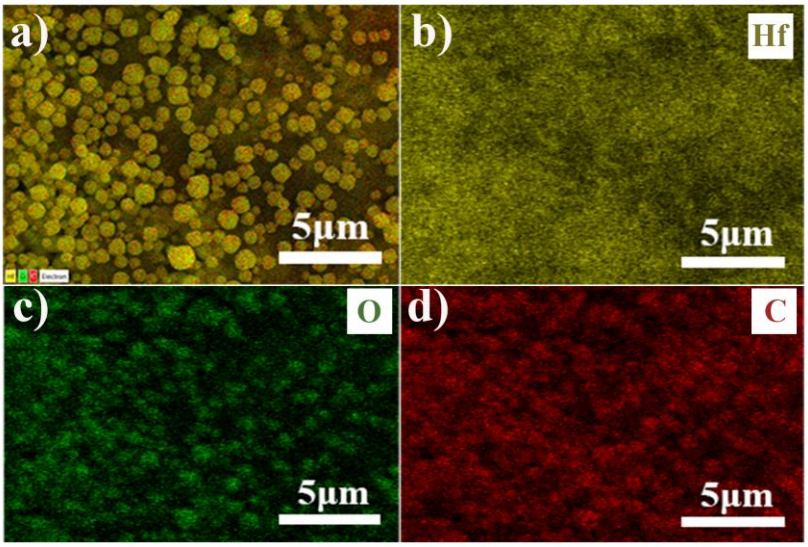

Fig. S8 EDS elemental analysis of the Li/HLMOF-4. a) EDS elemental mapping of all elements. EDS mapping of b) Hf element, c) O element and d) C element.

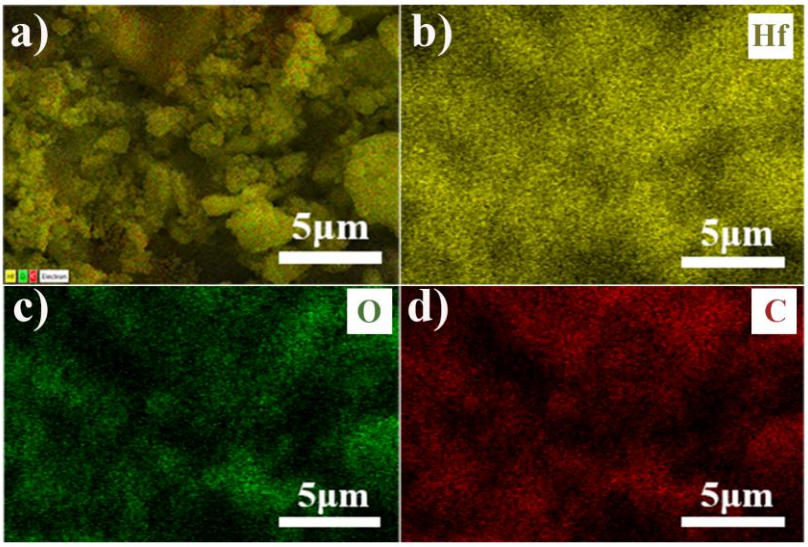

Fig. S9 EDS elemental analysis of the Li/HLMOF-5. a) EDS elemental mapping of all elements. EDS mapping of b) Hf element, c) O element and d) C element.

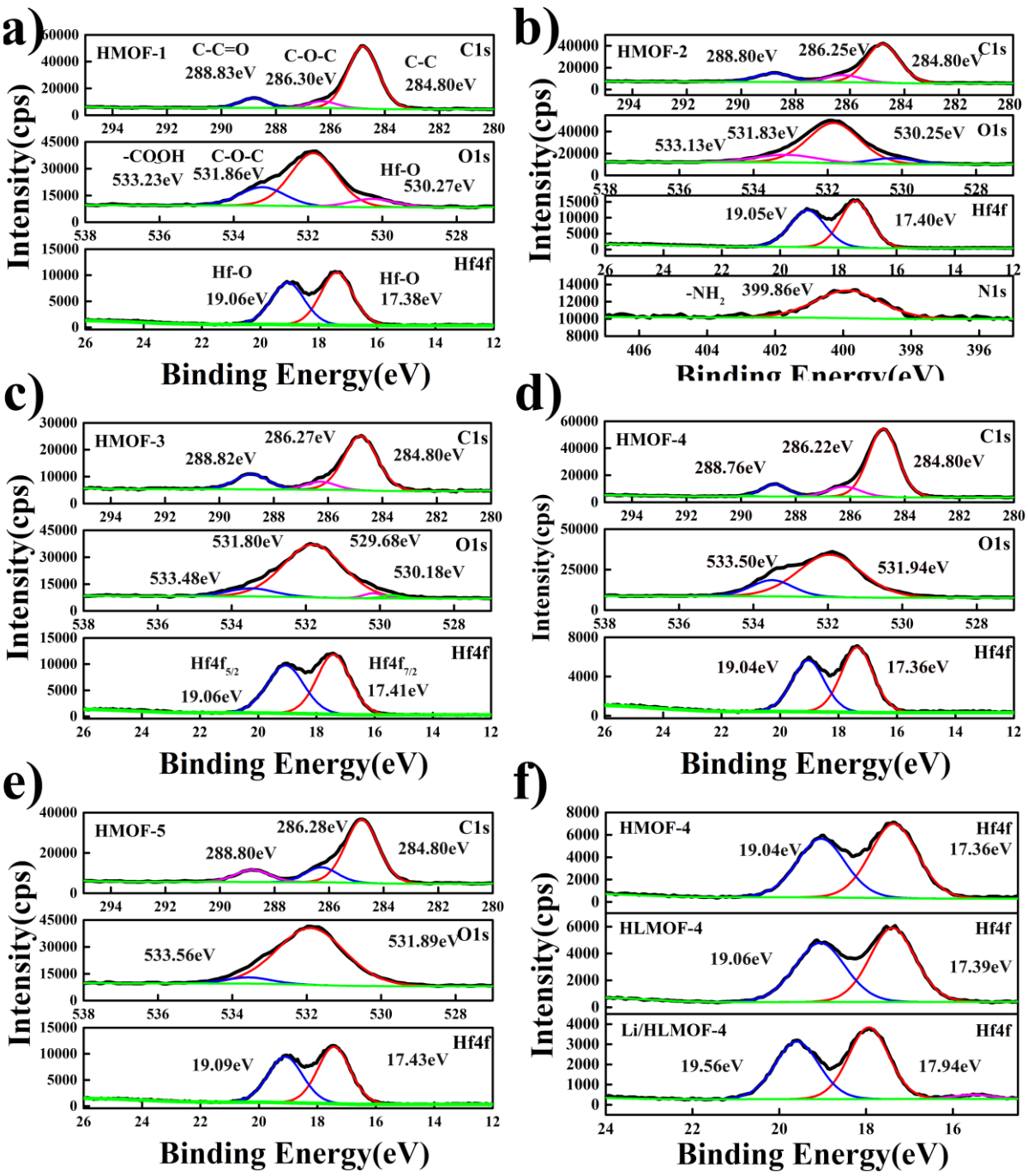

Fig. S10 XPS spectra of a) HMOF-1; b) HMOF-2; c) HMOF-3; d) HMOF-4; e) HMOF-5 and f) Hf 4f core-level spectrum of HMOF-4, HLMOF-4 and Li/ HLMOF-4.

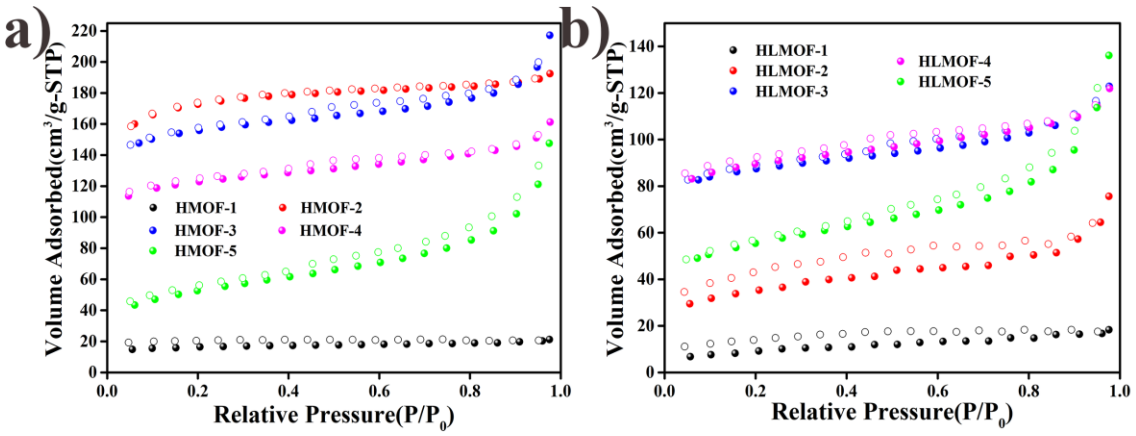

Fig. S11 N_2 adsorption- desorption profiles at 77 K for a) HMOFs and b) HLMOFs.

Table S1 BET of HMOFs and HLMOFs.

		Multi-point BET Surface Area m^2/g
HMOFs	HMOF-1	50.556
	HMOF-2	526.445
	HMOF-3	471.635
	HMOF-4	380.084
	HMOF-5	174.561
HLMOFs	HLMOF-1	32.946
	HLMOF-2	116.175
	HLMOF-3	265.439
	HLMOF-4	275.277
	HLMOF-5	178.575

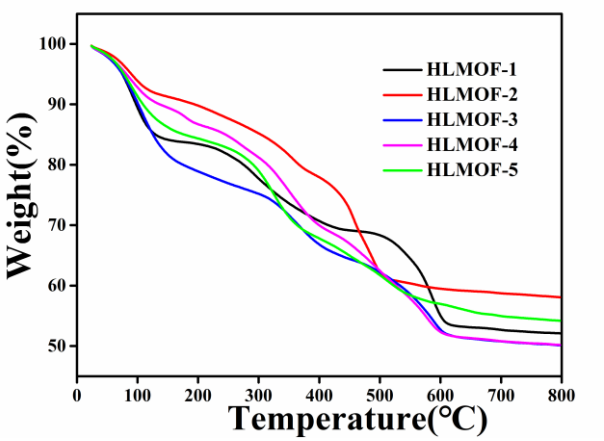

Fig. S12 TGA curves of the HLMOFs.

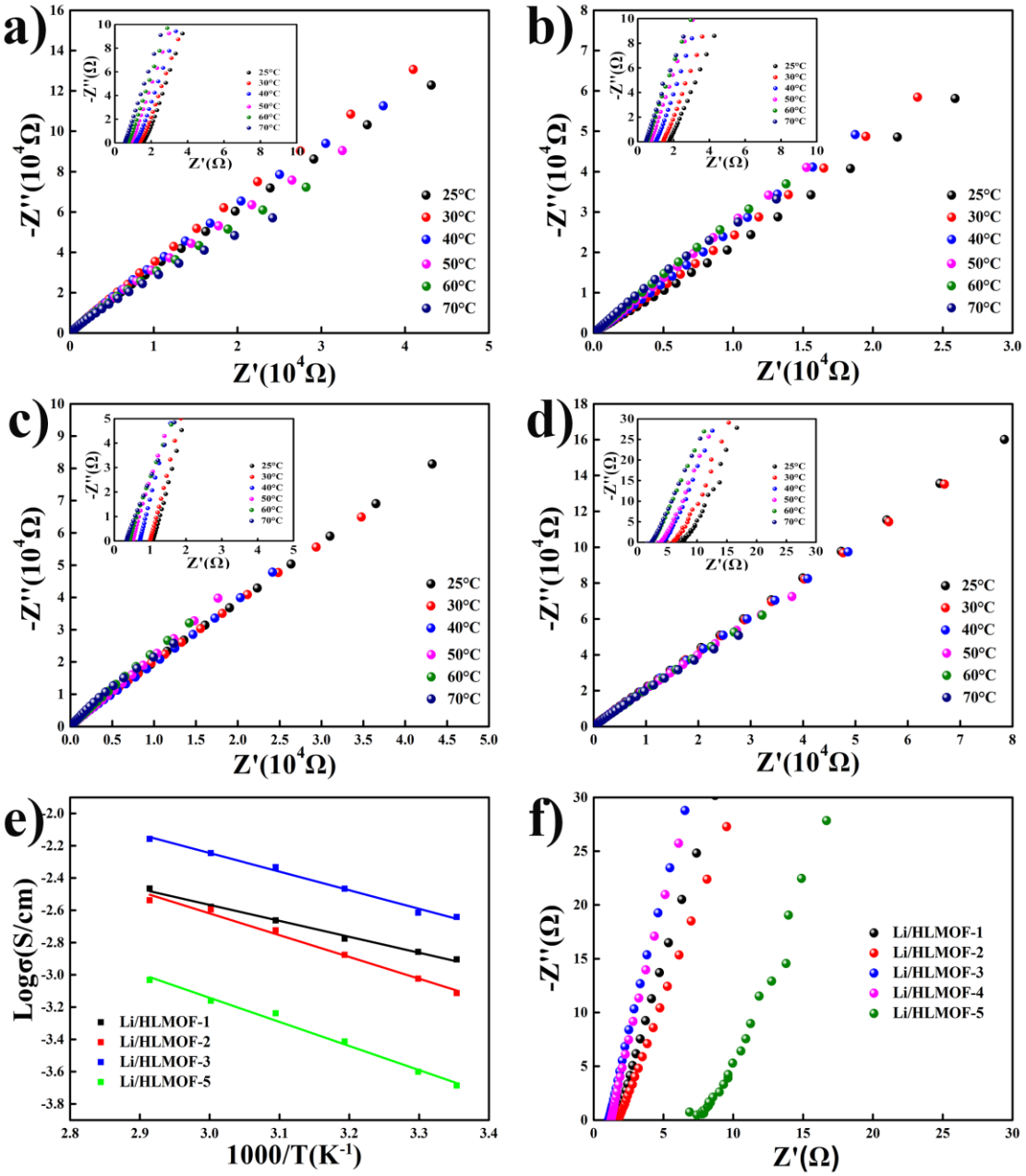

Fig. S13 EIS plots of a) Li/HLMOF-1 b) Li/HLMOF-2; c) Li/HLMOF-3; d) Li/HLMOF-5 electrolyte at temperatures from 25 °C to 70 °C; e) Arrhenius plots of electrolyte membrane; f) EIS plot of Li/HLMOFs at 25 °C.

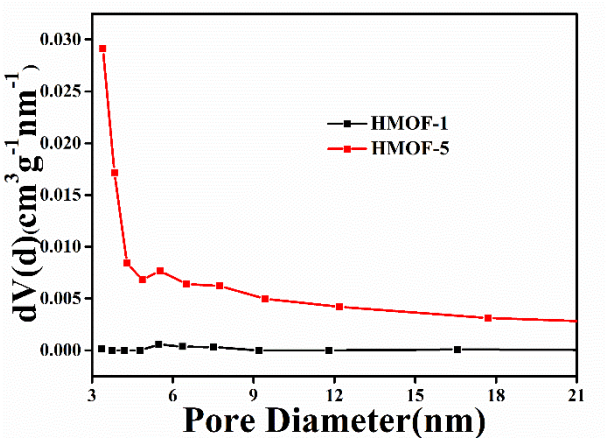

Fig. S14 The pore size distribution of HMOF-1 and HMOF-5.

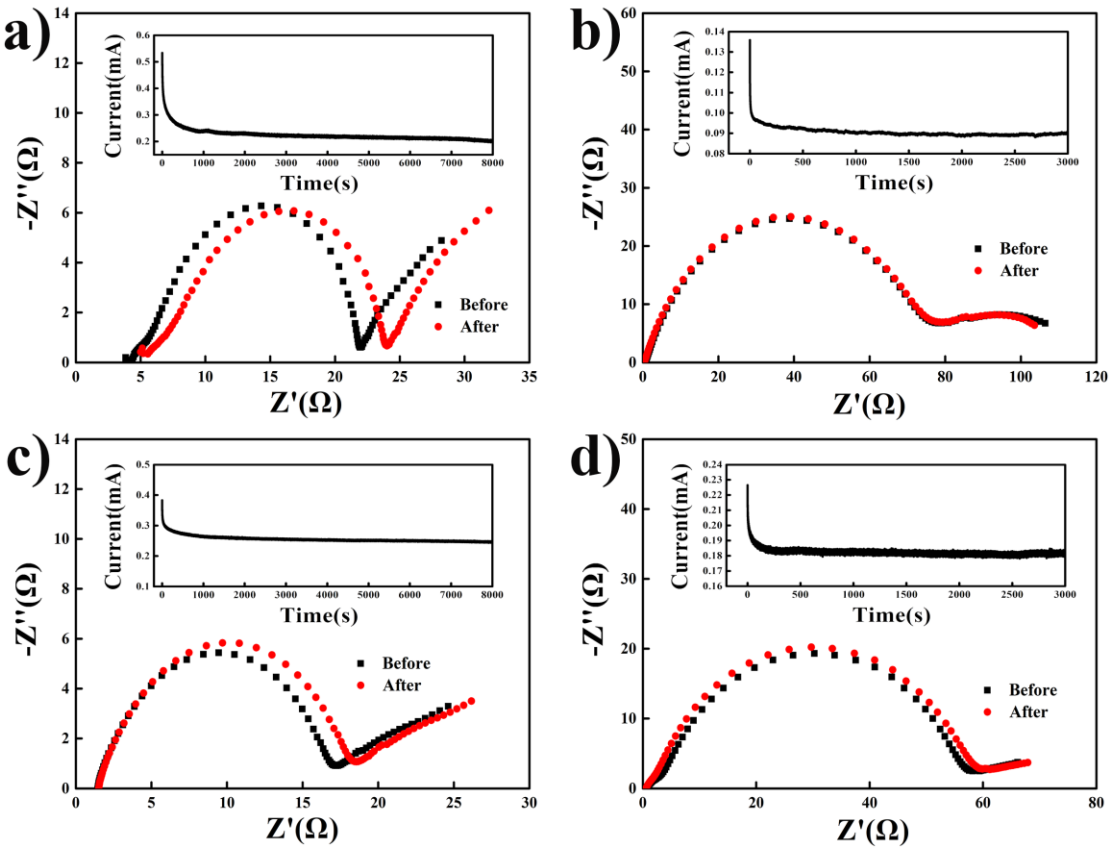

Fig. S15 EIS before and after polarization of the electrolyte of a) Li/HLMOF-1; b) Li/ HLMOF-2; c) Li/ HLMOF-3; d) Li/ HLMOF-5; the inset is current-time curve at 10 mV of polarization.

Table S2 Summary of solid-state electrolytes.

Electrolyte membrane	Ionic conductivity S/cm (25℃)	Potential window V	Ion transference number
Li/HLMOF-1	1.25×10^{-3}	1.50-4.55	0.20
Li/HLMOF-2	7.72×10^{-4}	1.40-4.45	0.19
Li/HLMOF-3	2.28×10^{-3}	1.50-4.62	0.44
Li/HLMOF-4	2.82×10^{-3}	1.60-4.65	0.58
Li/HLMOF-5	2.06×10^{-4}	1.85-3.80	0.15

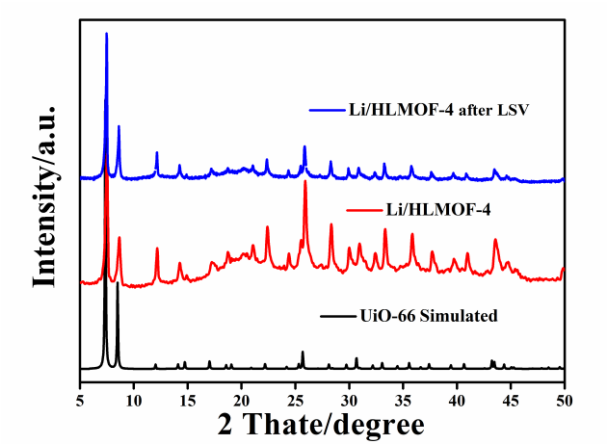

Fig. S16 The XRD of Li/HLMOF-4 electrolyte membrane before and after cycle.

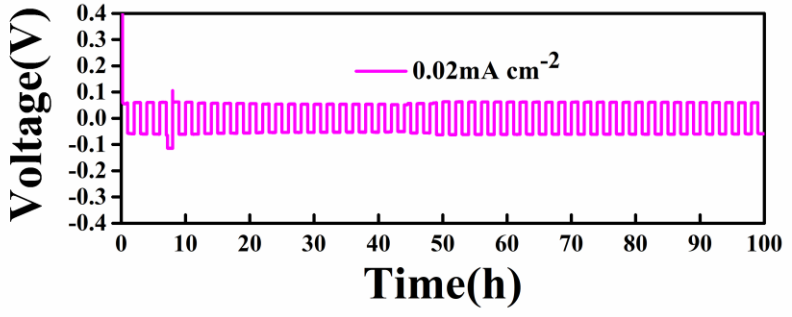

Fig. S17 The Li plating and stripping performance of the Li|| Li/HLMOF-4||Li cell at a current density of 0.02 mA cm^{-2}.

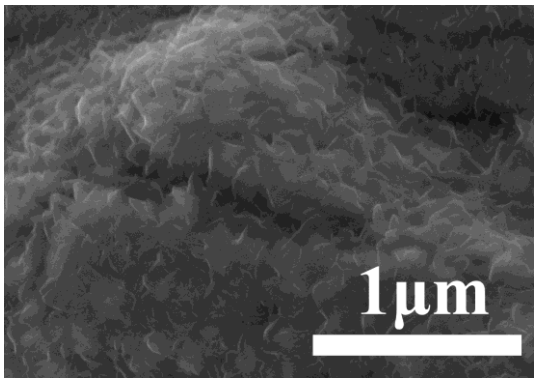

Fig. S18 SEM of lithium foil surface after plating/stripping experiments for Li|Li/HLMOF-4|Li symmetric cell.

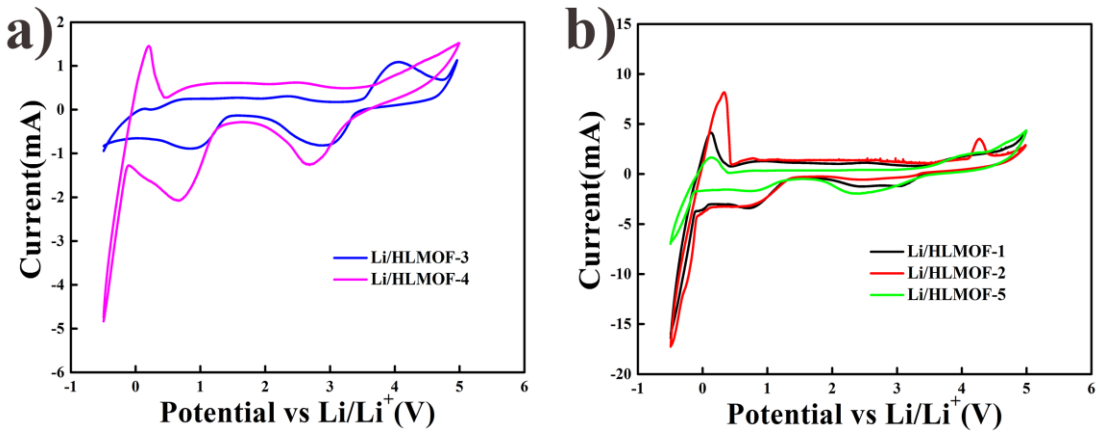

Fig. S19 The CV curve of a) Li/ HLMOF-3, Li/ HLMOF-4; b) Li/ HLMOF-1, Li/ HLMOF-2, Li/ HLMOF-5.