Energy Materials

Supplementary Material

Combining ternary, ionic liquid-based, polymer electrolytes with a single-ion conducting polymer-based interlayer for lithium metal batteries

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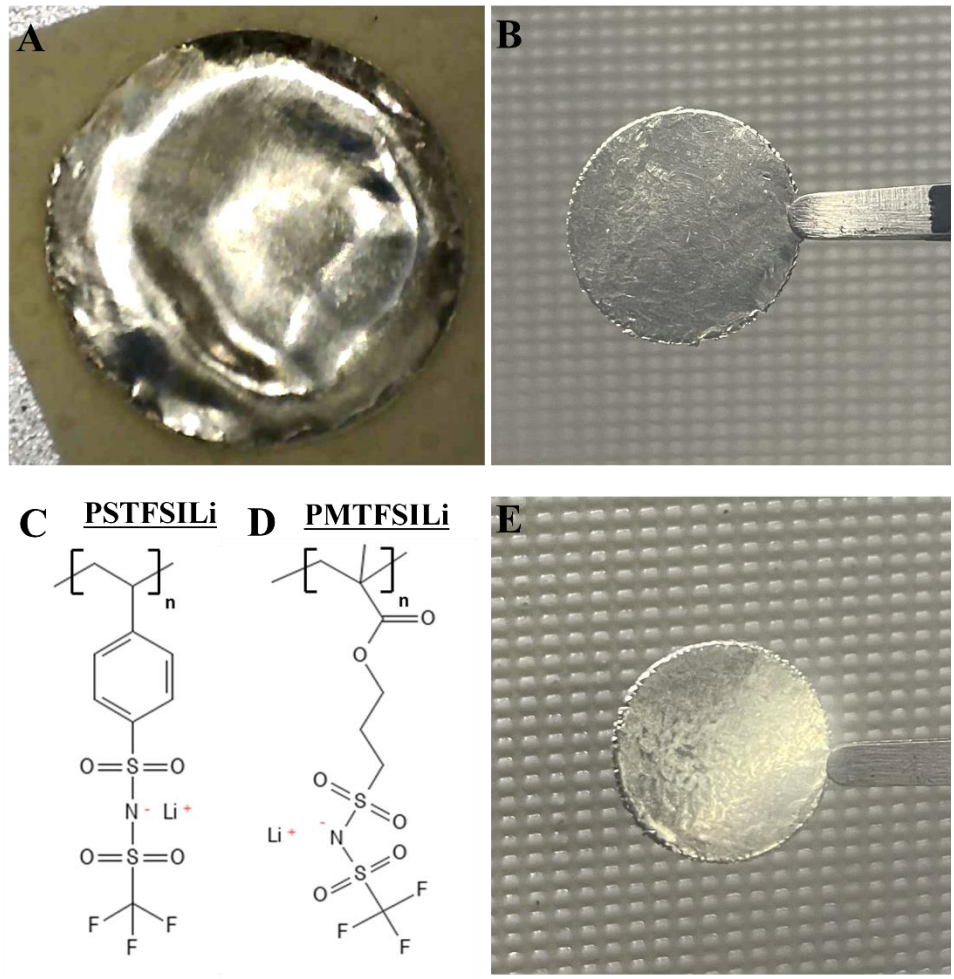
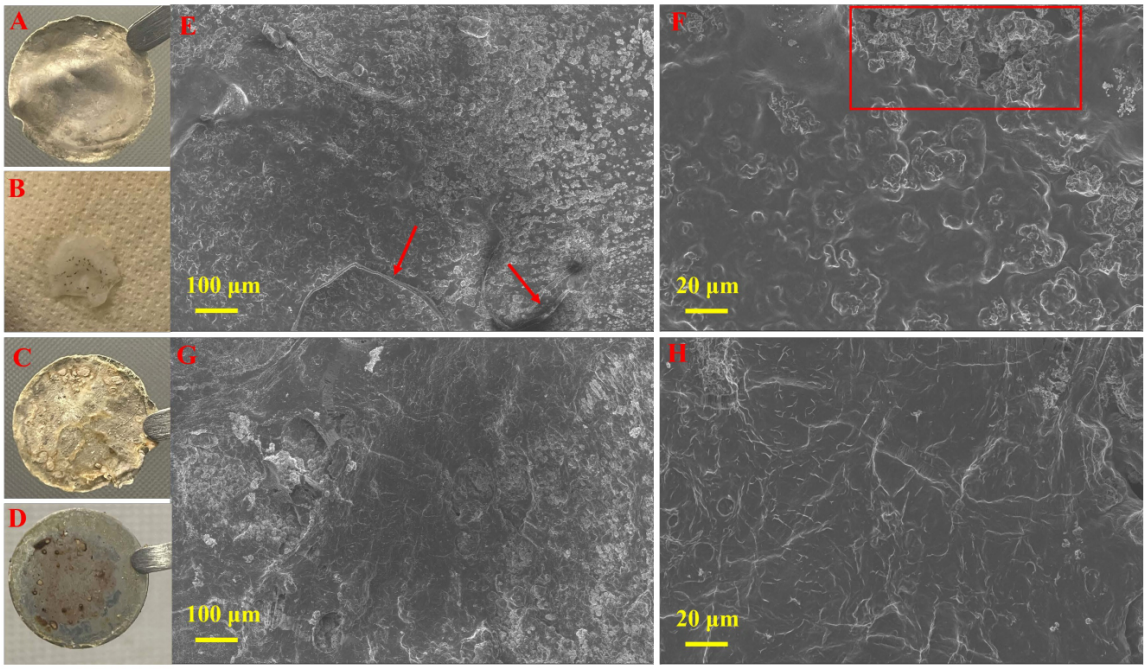
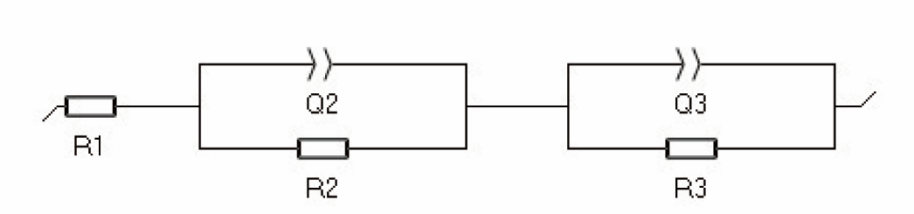


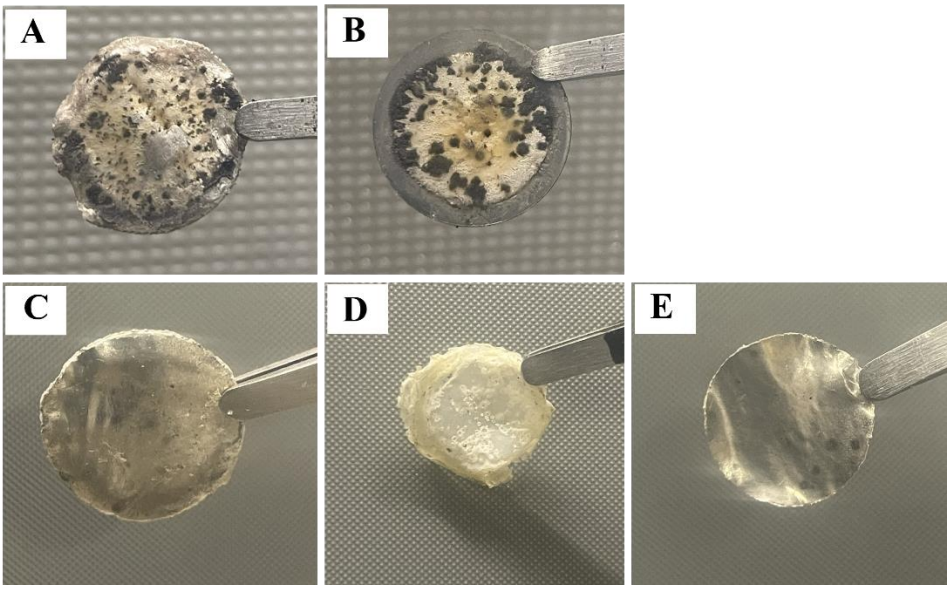
Supplementary Figure 1. (A) Photo of Li anode after polishing and rolling manually, (B) Photo of Li anode after pressing of (A), (C-D) chemical formula of PSTFSILi and PMTFSILi, (E) Li anode after coating of with 5wt.% PMTFSILi + 5 wt.% PSTFSILi /PC



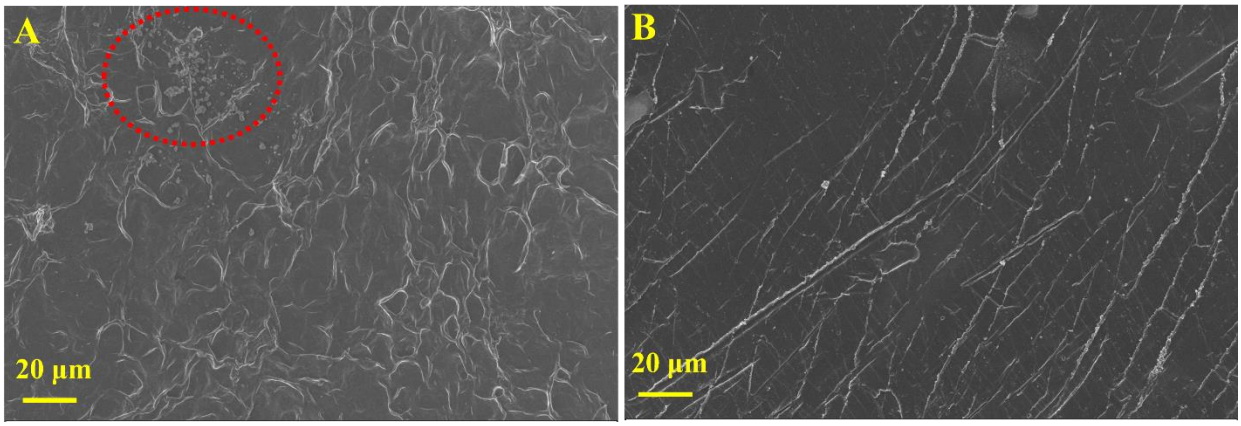
Supplementary Figure 2. (A) Photos of a plated *bare Li* electrode after Li electrodeposition at 0.1 mA cm^{-2} until short circuit, (B) corresponding TSPE after immersion in DME to help separate the Li electrode and the TSPE, (C-D) corresponding SEM images of the plated *bare* Li electrode. (E) Photos of a *coated Li* anode after Li electrodeposition at 0.1 mA cm^{-2} until Li depletion, (F) corresponding stainless steel spacer with almost no remain of the Li electrodissolved electrode, (G-H) corresponding SEM images of plated *coated* Li anode.



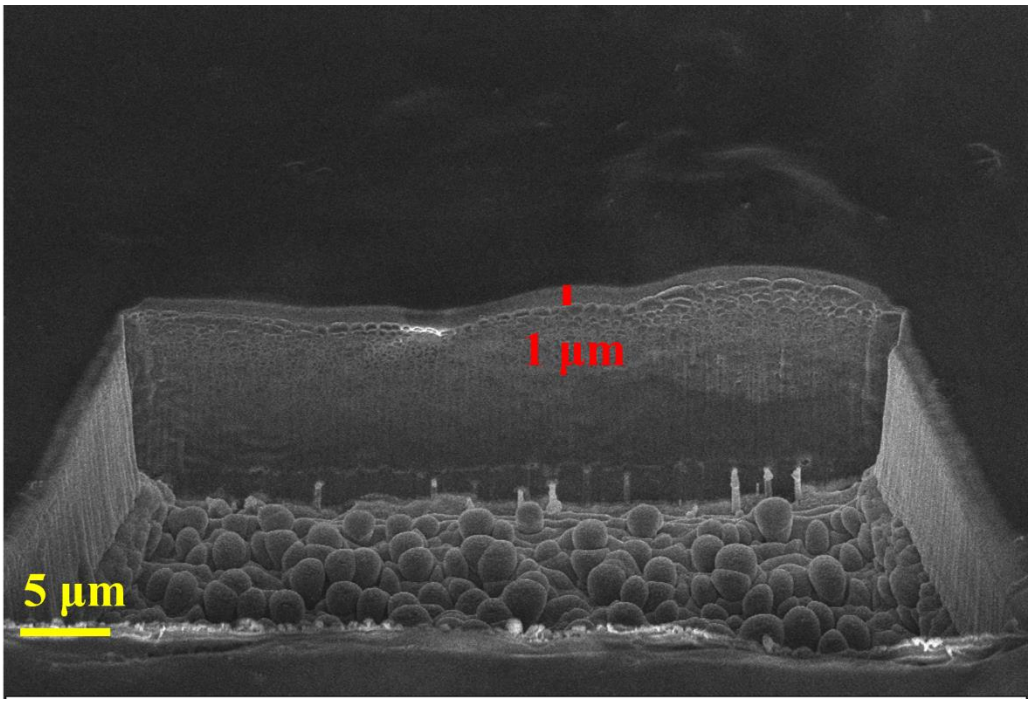
Supplementary Figure 3. Equivalent circuit for the fitting of the impedance spectra.



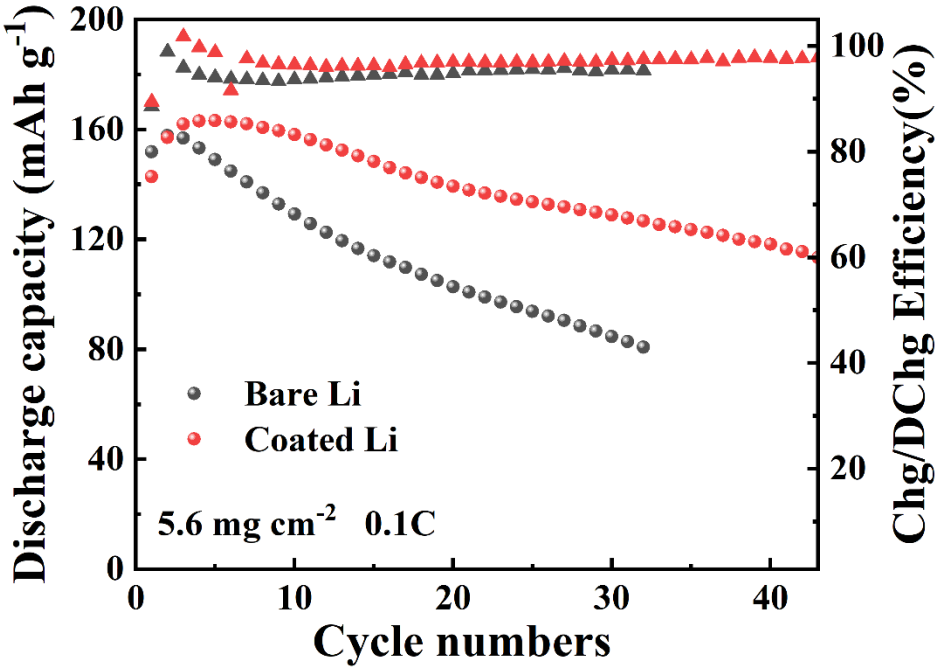
Supplementary Figure 4. Photos of Li|TSPE|Li cell components using *bare Li* (a-b) and *coated Li* (c-e) after 5 cycles at 0.1 mA cm^{-2} with 10h steps at 80 ℃. (A) initially plated Li anode, (B) initially stripped Li anode, (C) initially plated Li anode, (D) TSPE after immersion into DME and drying, (E) initially stripped Li anode.



Supplementary Figure 5. Top view of SEM images of (A) *bare Li* (B) and *coated Li* after 5 cycles at 0.1 mA cm^{-2} with 10h steps at 80 ℃.



Supplementary Figure 6. SEM images of *bare Li* after 5 cycles at 0.1 mA cm^{-2} with 10h steps at 80 ℃.



Supplementary Figure 7. Electrochemical performance of LFP|TSPE|Li cells cycling at 0.1C.

Supplementary Table 1. Resistances values obtained from fitting the impedance spectra.

		R_1/Ω	R_2/Ω	R_3/Ω	R_{total}/Ω
Bare Li	Rest 24	5.1	791.3	63.6	860.0
	1st cycle	4.4	483.7	32.4	520.6
	5th cycle	4.6	435.9	19.8	460.3
Coated Li	Rest 24	4.3	295.7	129.4	429.4
	1st cycle	3.5	280.4	74.2	358.1
	5th cycle	3.7	234.8	106.7	345.2

Supplementary Table 2. Surface atomic concentrations of elements for different Li electrodes.

	C	O	Li	F	S	N	Si	Cl
Pristine bare Li	26.4%	32.0%	34.0%	-	-	-	7.6%	-
Pristine coated Li	41.5%	24.8%	18.0%	4.9%	2.7%	2.0%	5.7%	0.4%
Bare Li-cycled	54%	18.5%	21.5%	4.0%	0.3%	0.5%	0.3%	0.8%
Coated Li -cycled	66.8%	17.1%	11.1%	3.4%	0.6%	0.3%	0.4%	0.4%

Supplementary Table 3. XPS fitting results of C 1s spectra for samples.

		C-C/C-H/C=C	C-O	O-C=O	CF_x
Pristine bare Li	Binding energy /eV	284.8	286.7	289.2	-
	Content	22.2%	1.8%	2.5%	-
Bare Li-cycled	Binding energy /eV	284.8	286.4	288.8	-
	Content	40.5%	7.1%	6.4%	-
Pristine coated Li	Binding energy /eV	284.8	286.6	289.1	292.7
	Content	28.0%	5.4%	6.6%	1.6%
Coated Li-cycled	Binding energy /eV	284.8	286.7	288.4	-
	Content	46.5%	14.4%	5.9%	-

74 **Supplementary Table 4.** XPS fitting results of O 1s spectra for samples.

		LiO_2	O-C=O	O-C/O=S
Pristine bare Li	Binding energy /eV	528.6	531.6	
	Content	6.0%	26.0%	
Bare Li-cycled	Binding energy /eV	528.4	531.6	533.0
	Content	1.1%	16.2%	1.3%
Pristine coated Li	Binding energy /eV	-	532.0	533.0
	Content	-	19.5%	5.2%
Coated Li-cycled	Binding energy /eV	-	531.5	533.1
	Content	-	9.0%	8.1%

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76 **Supplementary Table 5.** XPS fitting results of Li 1s spectra for samples.

		Li_2O	Li^+
Pristine bare Li	Binding energy /eV	53.9	55.2
	Content	15.2%	18.8%
Bare Li-cycled	Binding energy /eV	53.8	55.2
	Content	4.0%	17.5%
Pristine coated Li	Binding energy /eV	-	55.4
	Content	-	18.0%
Coated Li-cycled	Binding energy /eV	-	55.4
	Content	-	11.1%

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79 **Supplementary Table 6.** XPS fitting results of F 1s spectra for samples.

		LiF	CF_x
Pristine bare Li	Binding energy /eV	-	-
	Content	-	-
Bare Li-cycled	Binding energy /eV	685.0	688.2
	Content	3.2%	0.8%
Pristine coated Li	Binding energy /eV	684.9	688.6
	Content	1.5%	3.4%
Coated Li-cycled	Binding energy /eV	684.9	-
	Content	3.4%	

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