Supporting information

In-situ Polymerization of Dicationic Pyrrolidinium-based Solid Polymer Electrolyte for High Performing and Long-Lasting Lithium-Ion Battery

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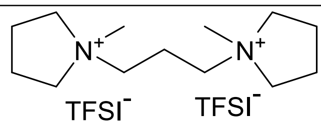
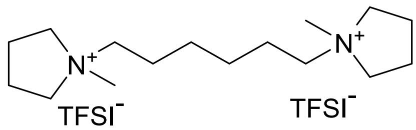
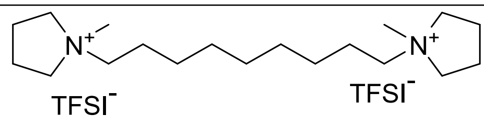
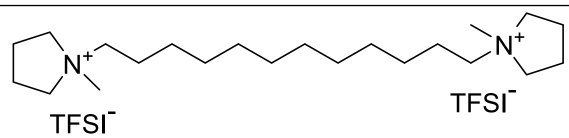
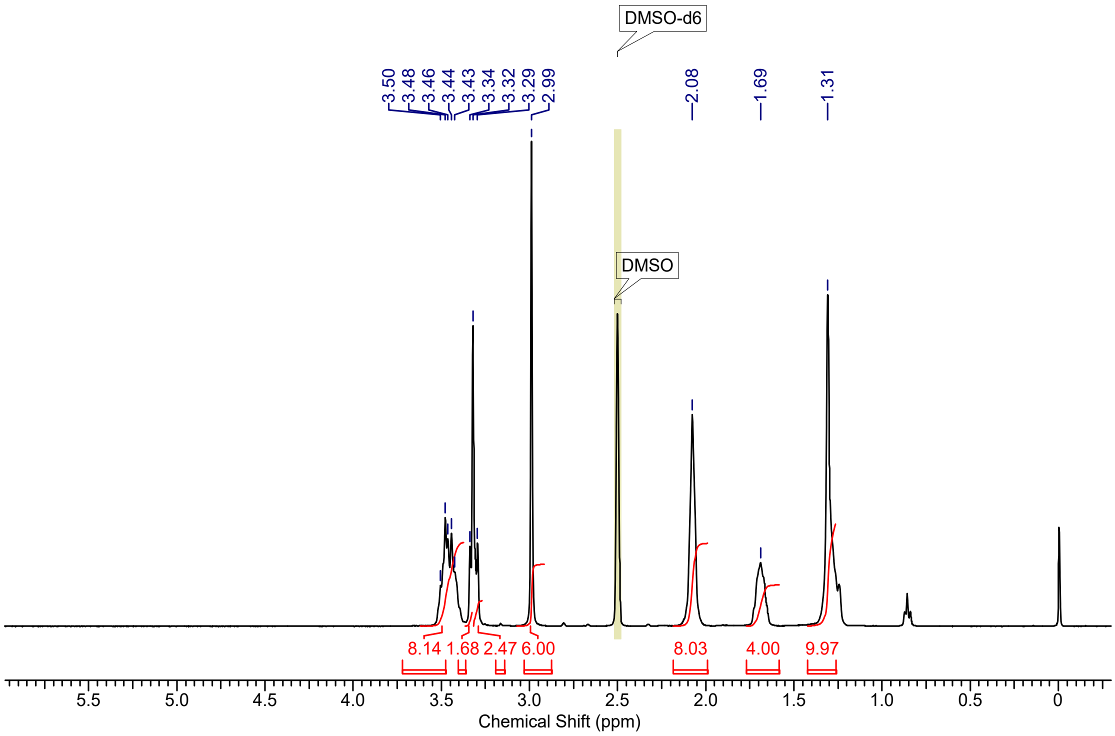
Table S1: Abbreviated forms of the DILs

Sr. No	Abbreviations	Name of Ionic Liquids	Structures
1	$[C_3(pyr)_2]$ $[TFSI]_2$	1,3-Bis (1-methyl pyrrolidinium 1-yl) propane bis (trifluoro methyl sulfonyl) imide	
2	$[C_6(pyr)_2]$ $[TFSI]_2$	1,6-Bis (1- methyl pyrrolidinium 1-yl) hexane bis (trifluoro methyl sulfonyl) imide	
3	$[C_9(pyr)_2]$ $[TFSI]_2$	1,9-Bis (1- methyl pyrrolidinium 1-yl) nonane bis (trifluoro methyl sulfonyl) imide	
4	$[C_{12}(pyr)_2]$ $[TFSI]_2$	1,12-Bis (1- methyl pyrrolidinium 1-yl) dodecane bis (trifluoro methyl sulfonyl) imide	



^{1}H NMR (400 MHz, δ/ppm): 3.28–3.32 (m, 4H, $N^+–CH_2–(CH_2)_7–CH_2–N^+$), 3.18–3.26 (m, 8H, pyrrolidinium $N^+–CH_2$), 1.65–1.77 (m, 8H, pyrrolidinium $–CH_2–$), 1.23–1.35 (m, 4H, linker $–CH_2–CH_2–N^+$), 1.20–1.32 (m, 10H, linker internal $–(CH_2)_5–$).

Figure S1: ^{1}H NMR spectrum of the synthesized dicationic ionic liquid $[C_9(pyr)_2][TFSI]_2$ recorded in $DMSO-d_6$ as the solvent.

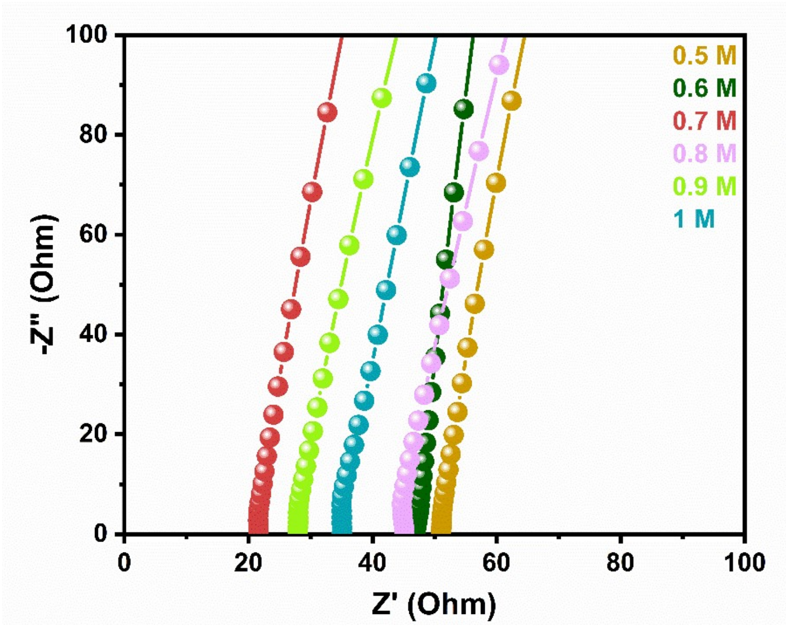


Figure S2: Electrochemical impedance spectroscopy (EIS) Nyquist plots of LITFSI-DIL electrolyte with varying salt concentration (0.5M-1.0M).

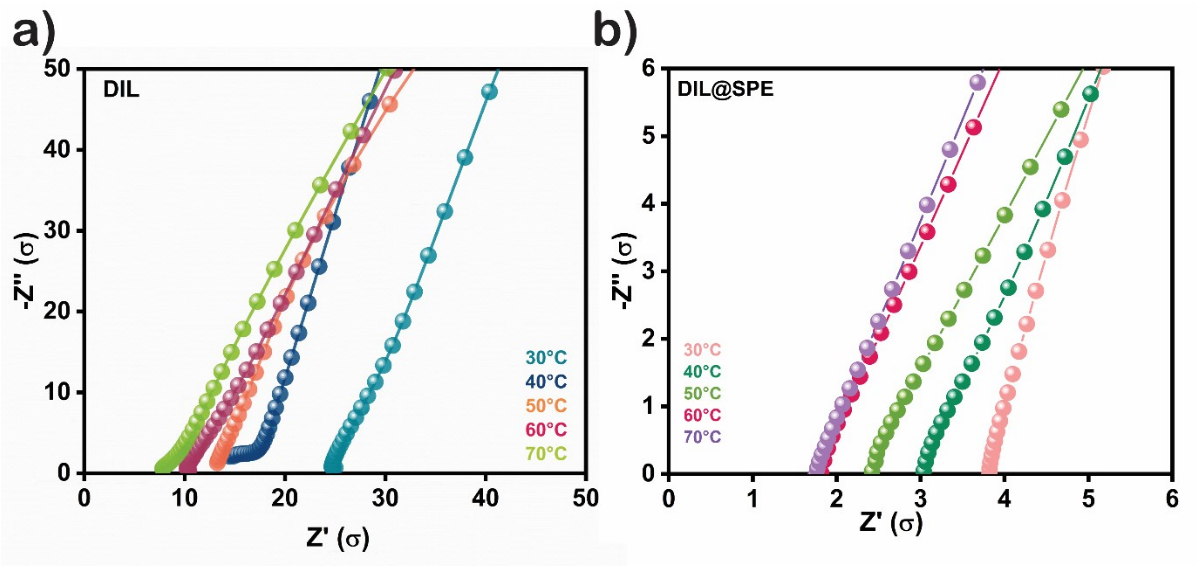


Figure S3: Temperature-dependent electrochemical impedance spectroscopy (EIS) Nyquist plots of a) DIL and b) DIL@SPE systems measured in the temperature range of 30-70 °C.

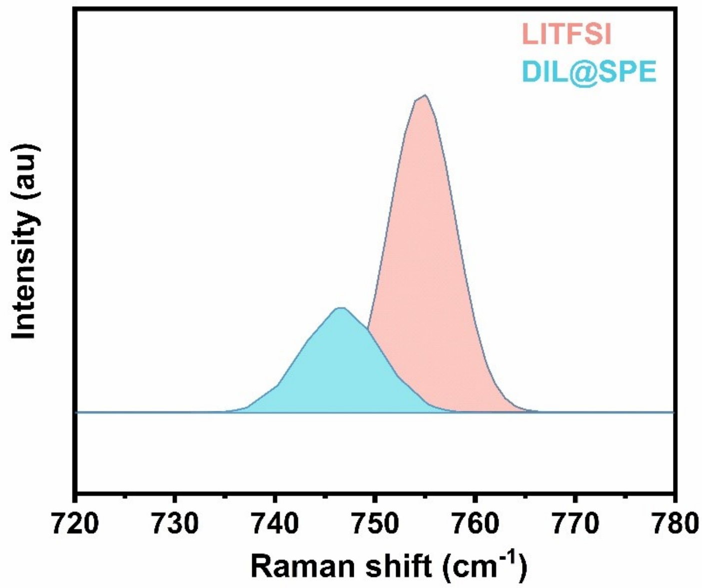


Figure S4: Raman spectra of pure LiTFSI and the DIL@SPE electrolyte

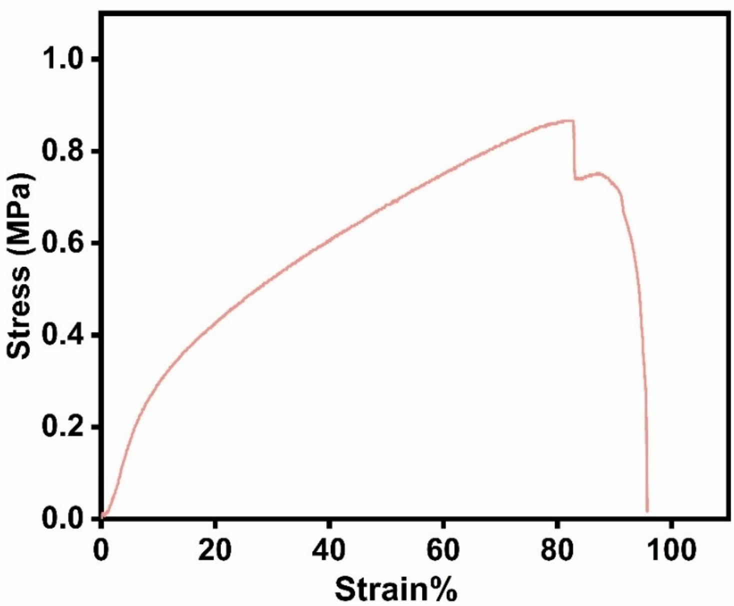


Figure S5: Tensile stress-strain curves of the solid polymer electrolyte membranes obtained from mechanical testing

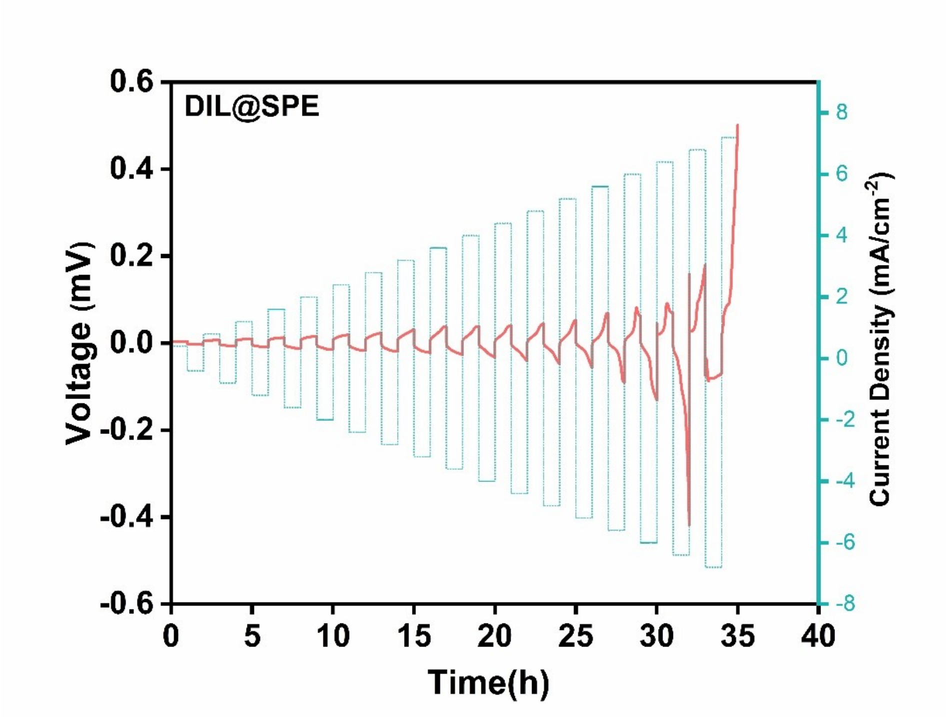


Figure S6: Critical current density (CCD) test of the symmetric Li|DIL@SPE|Li at 30 °C.

TableS2: HOMO, LUMO, and Chemical Hardness (*η*)

Molecule	HOMO(eV)	LUMO(eV)	Energy Gap(eV)	Chemical hardness η (eV)	Interpritation
LiTFSI	-8.91	-1.708	7.202	3.60	Reactive; SEI formation
VEC	-8.135	-1.024	7.111	3.56	Moderately reactive
ETPTA	-7.083	-1.756	5.327	2.66	Crosslinker, good reactivity
DIL	-6.609	-0.284	6.325	3.16	Stable, electrochemically robust

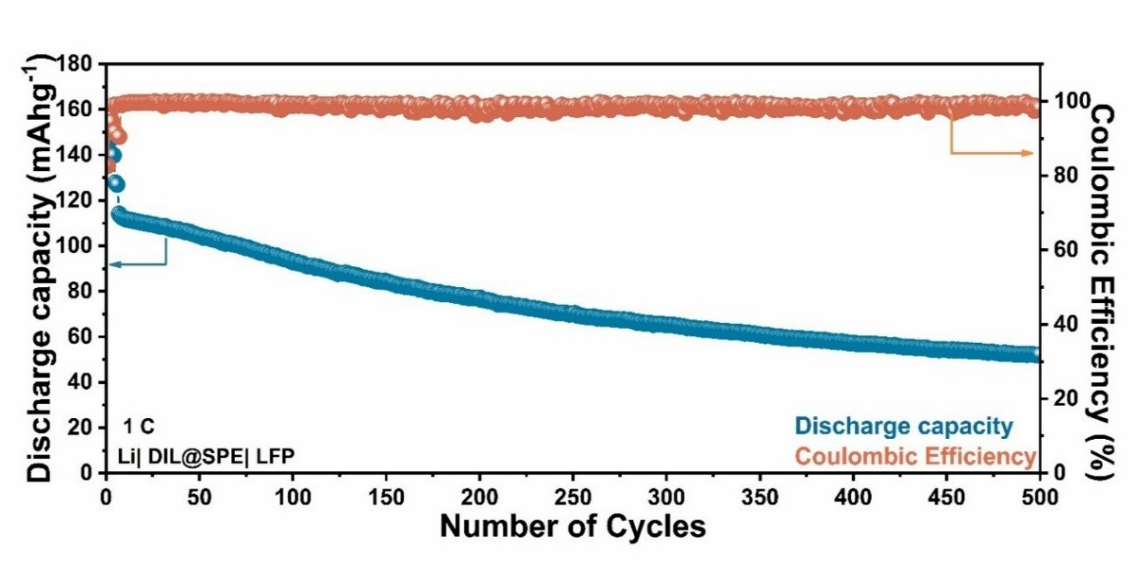


Figure S7: Long-term cycling performance of the LFP|DIL@SPE|Li cell at a 1C rate

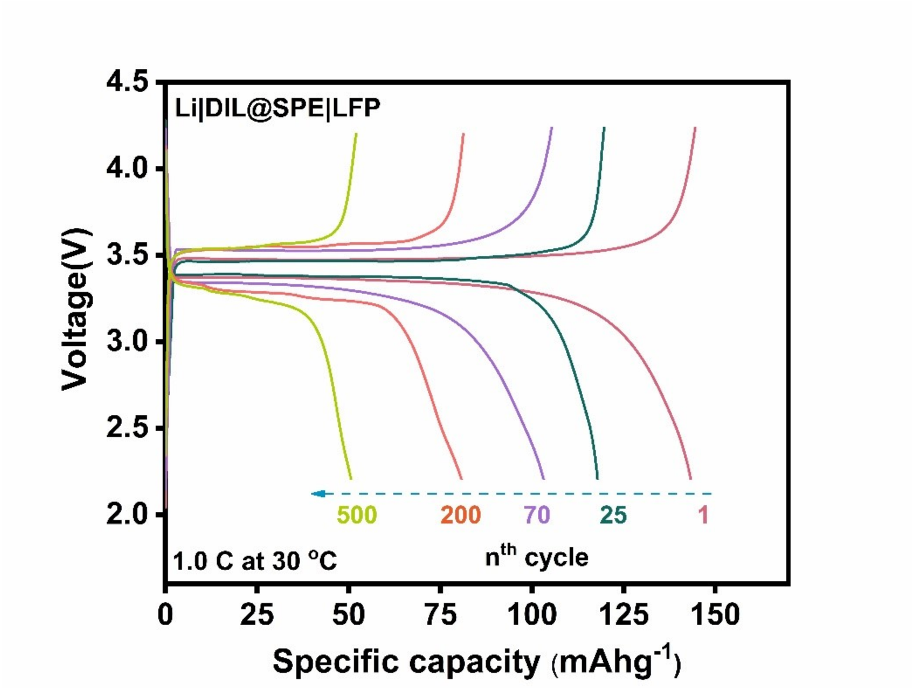
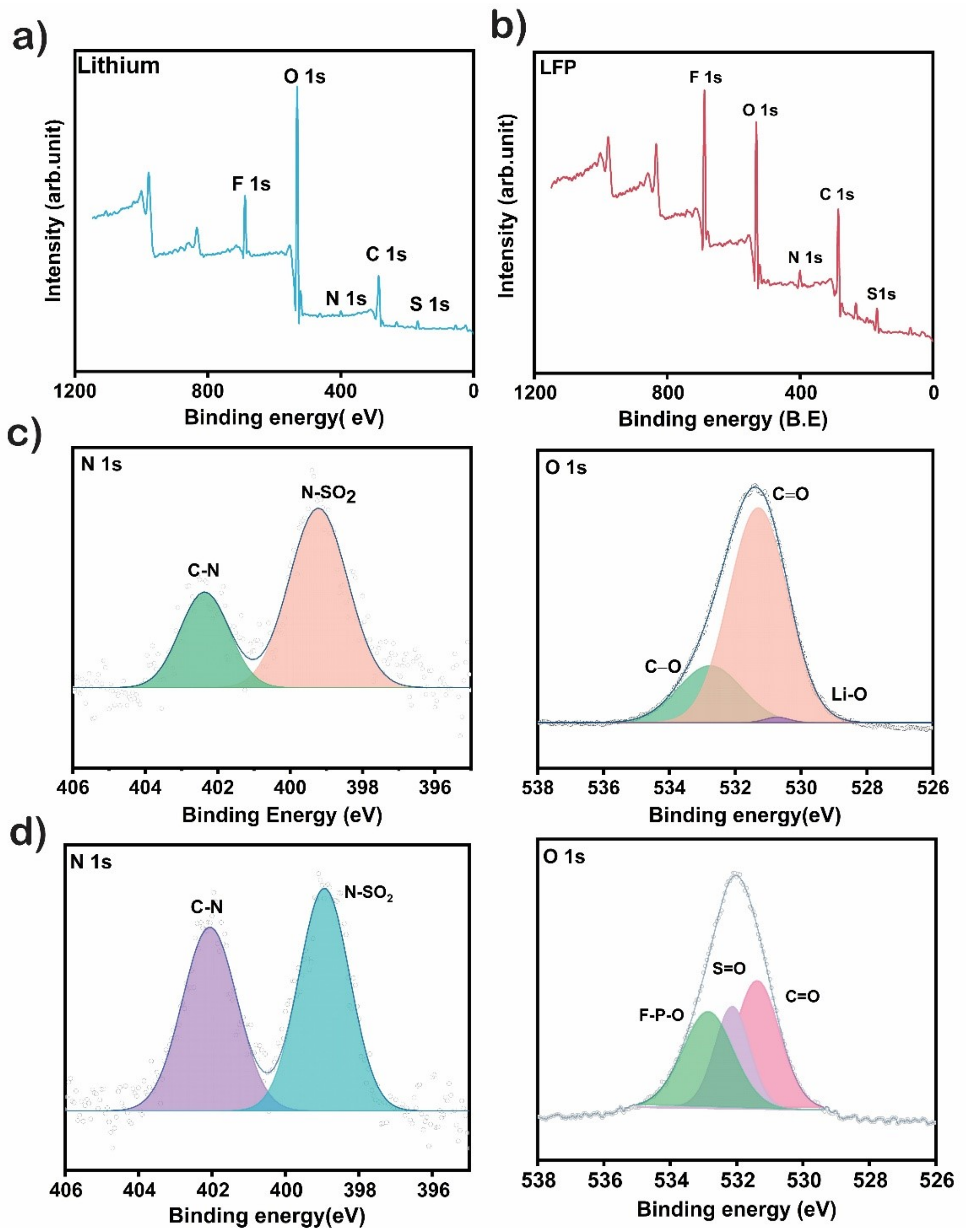


Figure S8: Galvanostatic charge-discharge profile of the LFP |DIL@SPE | Li cell various cycles at 1C.



Figur S9: Post-mortem XPS spectra of a) Li anode and b) LFP cathode from LFP|DIL@SPE|Li cell after 100 cycles at 1C, c) N1s and O 1s peaks of lithium anode d) N1s and O 1s peaks of LFP cathode

Table S3: comparative literature data on the reported system

Configuration	**C-rate and corresponding specific capacity [mAh g_{-1}]**					**Number of cycles**	**Reference Component**
	0.05C	**0.1C**	**0.2C**	**0.5C**	**1C**		
Li/IPN-GPE/$LiFePO_4$	-	152.1	146	132	107.6	100 @ 1C RT	[1]
LFP/PIL-SPE-LiTFSI-40/Li	-	164.3	153.1	121.2	95.7	100@ 0.5C RT	[2]
LFP|0.6 M LiTFSI/$[C_6(mim)_2]$ [TFSI]2|Li	134	133	113.3	73.6	-	100@1C RT	[3]
LFP |Gel electrolyte| Li	166	164	164	143	128	130 @1C RT	[4]
LFP/PIL-LiTFSI-LATP/Li	-	163.2	160.2	152.5	138.4	250 @1C 60^{o}C	[5]
Li/NPE-V2/LiFePO4		158.2	152.6	141.5	119.4	100@0.1C &0.5C	[6]
Li/P(PEGMA-VBImTFSI2)/LiTFSI/ LiFePO4	-	142.2	-	-	-	100 @0.1C 25^{o}C	[7]
LiFePO4|PIL-QSE|Li	135	128	116	104	97	100@ 0.1C 25^{o}C	[8]
Li/XP-20/$LiFePO_4$	-	168.2	160.3	134.1	70.0	150 @ 0.2C 25^{o}C)	[9]
LFP/GPE/Li	-	150.04	151.02	141.44	132.02	200@0.5C	[10]
Li/PILGPE@IL/LiFe O4		152	148	144	129	120@0.5C	[11]
PIL. Li/$LiFePO_4$	-	153.7	-	-	-	150@0.1C 25 °C.	[12]
Li |0.2M LiTFSI-[ME3AMIm] [TFSI] | LiFePO4	98.5	67.4	35.4	-	-	30@0.05C RT	[13]
Li|MPILelectrolyte| $LiFePo_4$		137.4	127	112.6	110	100@ 1C 25 °C	[14]
Li/IPN EM/LiFePO4	110	85	82	-	76	200@ 1C RT	[15]
✓ **In this work, the electrochemical cycling tests were carried out for 500 cycles at 0.5 C & 1C**							
Li/DIL@SPE /LiFePO4		**164**	**150**	**127**	**81**	**500@0.5C & 1C RT**	**This Work**

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