Supplementary information

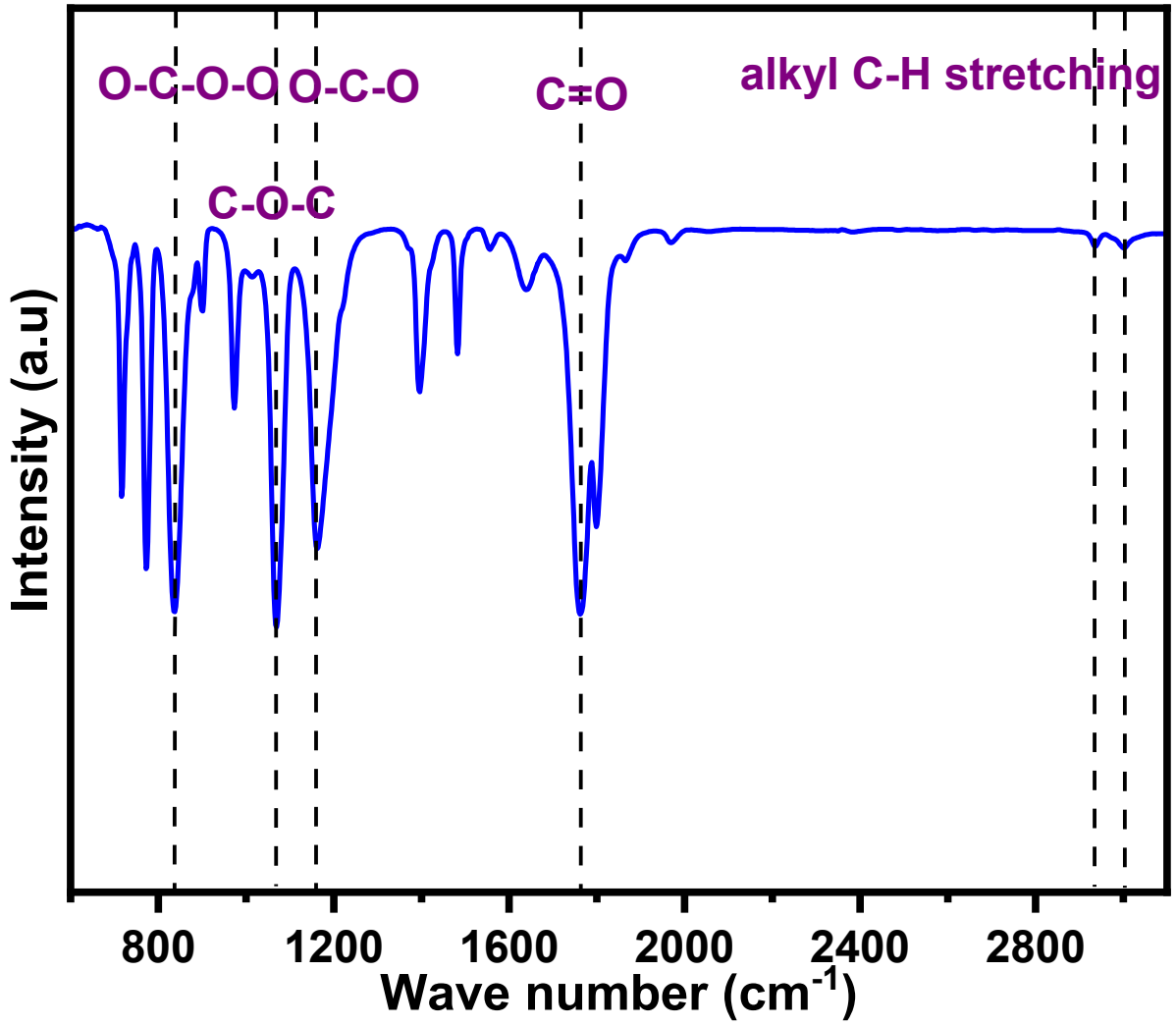


Figure S1 IR spectra showing the peaks corresponding to PEC in G-SPE.

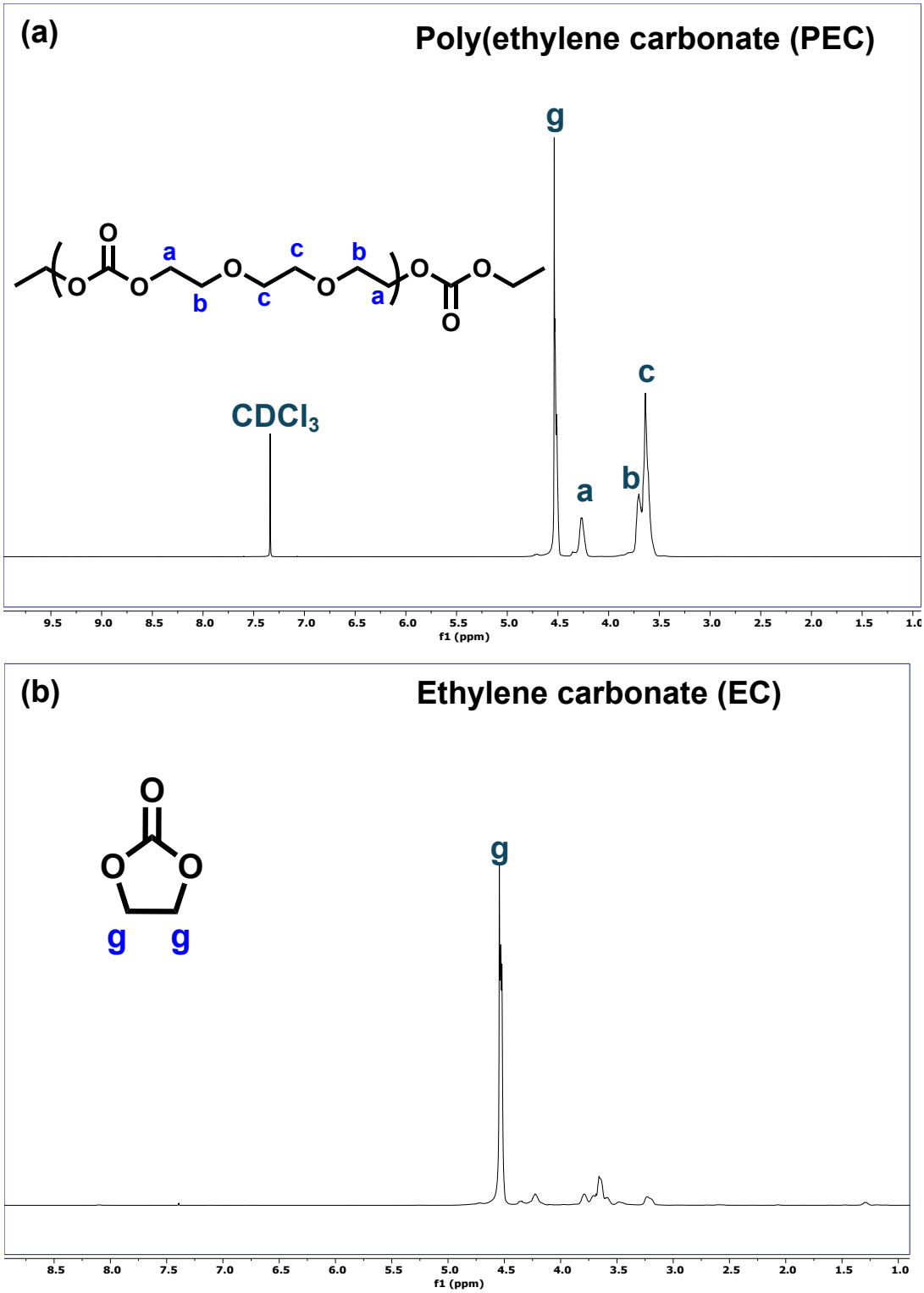
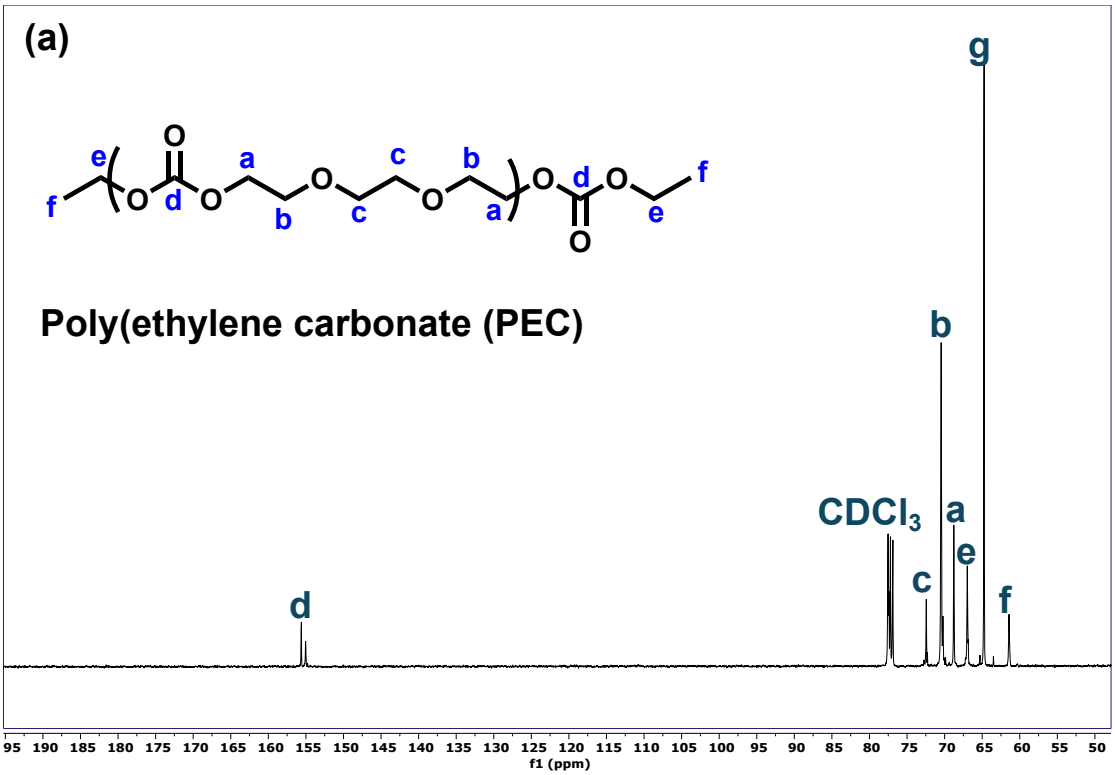


Figure S2 ^{1}H NMR spectra of (a) Poly(ethylene carbonate) (PEC), and (b) Ethylene carbonate (EC).



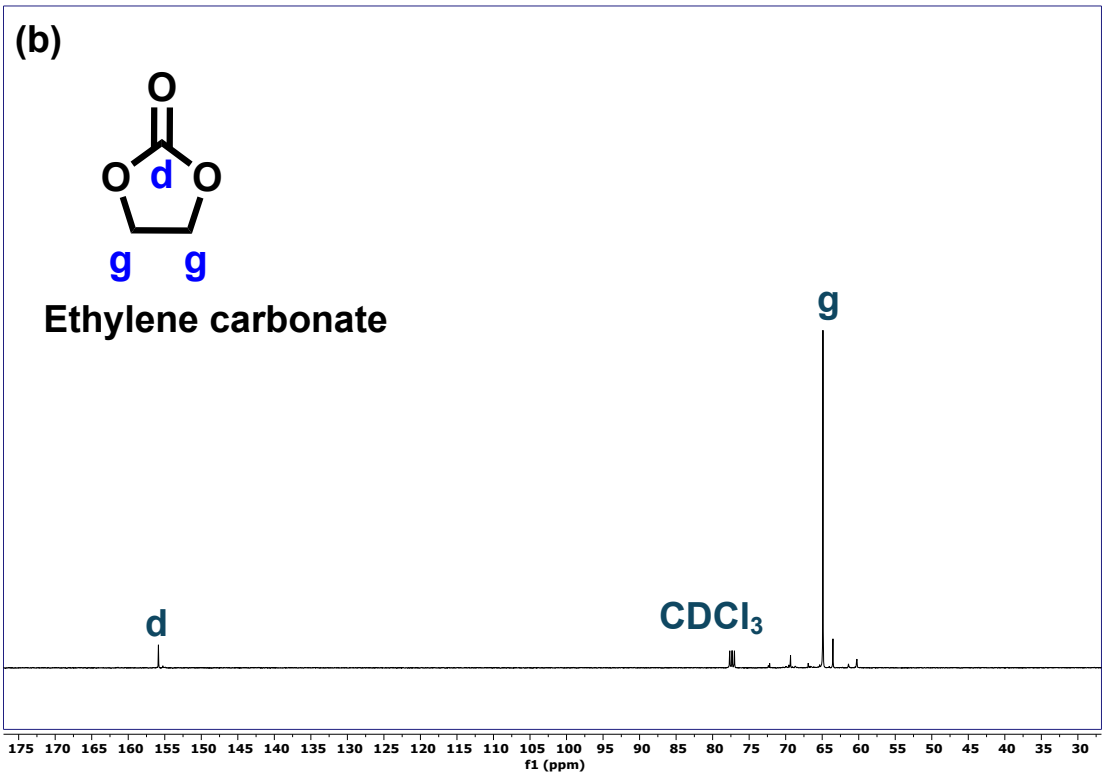


Figure S3 ^{13}C NMR spectra of (a) Poly(ethylene carbonate) (PEC), and (b) Ethylene carbonate (EC).

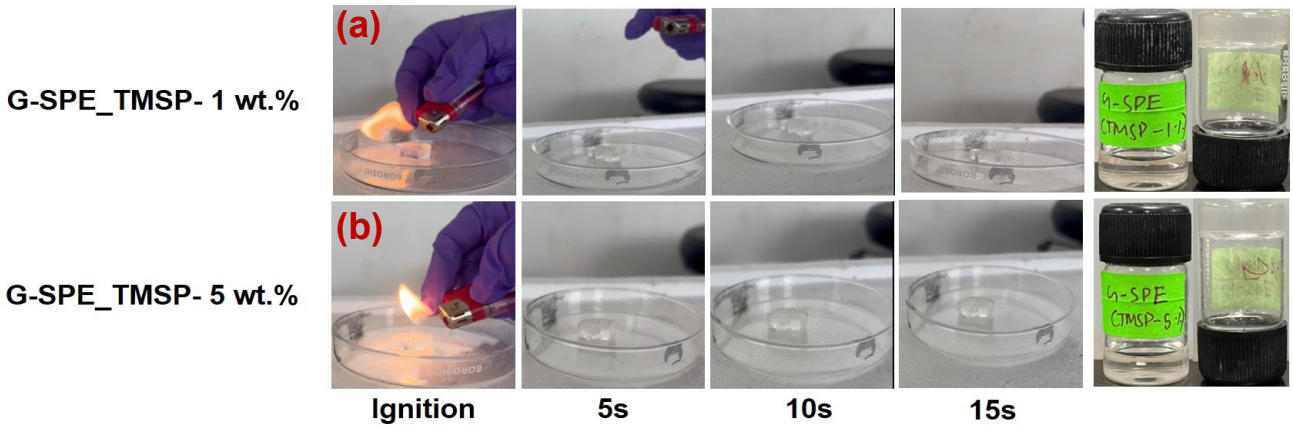


Figure S4 Flammability test of G-SPEs with TMSP concentrations of- (a) 1 wt.%, (b) 5 wt.% at durations of 5, 10, and 15s.

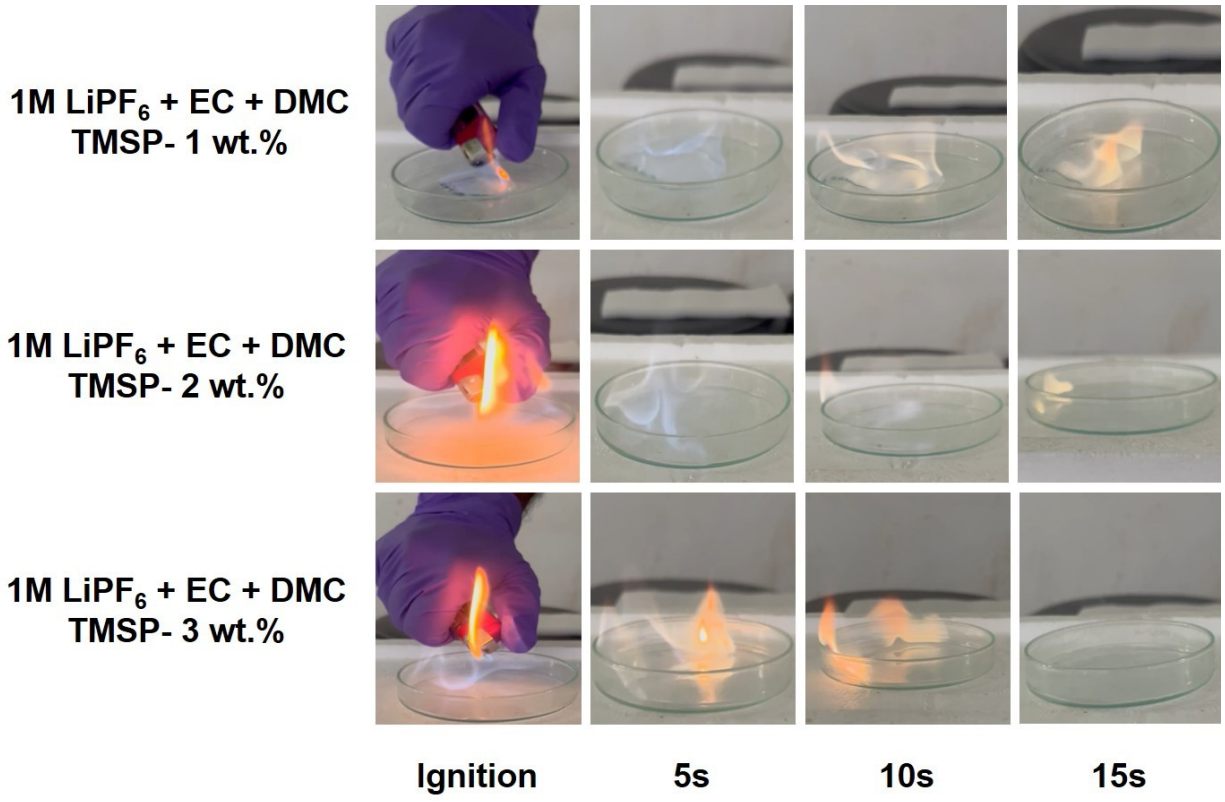


Figure S5 Flammability test of liquid electrolytes: 1M $LiPF_6$ + EC + DMC + TMSP- (a) 1 wt.%, (b) 3 wt.%, and (c) 5 wt.% at durations of 5, 10, and 15s.

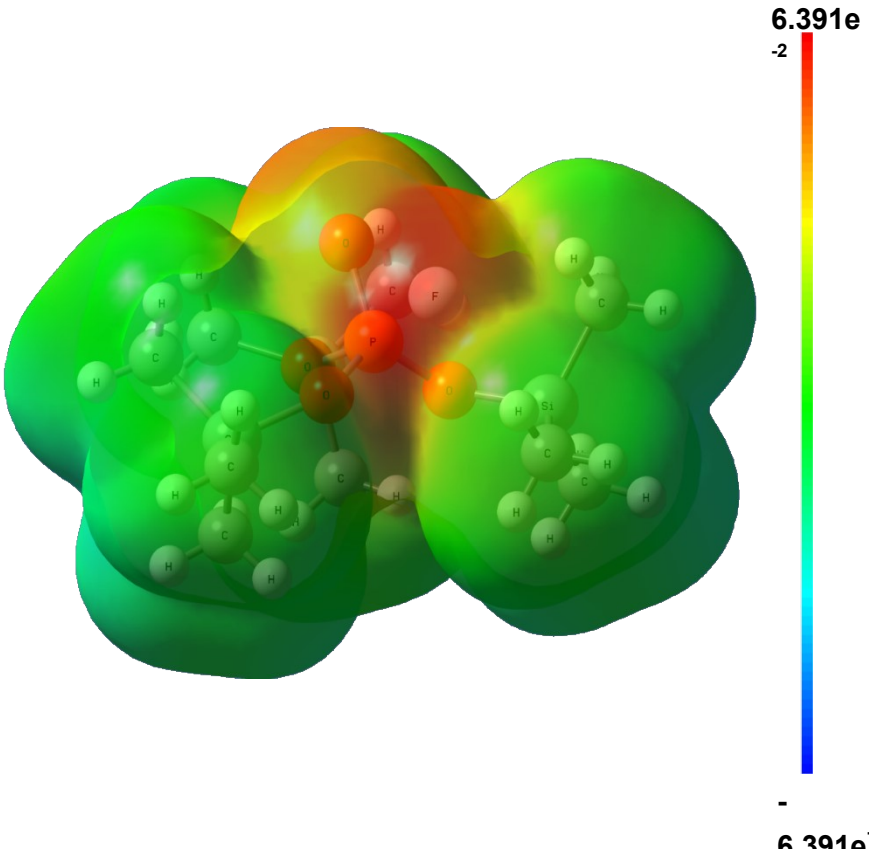


Figure S6 Shows the electrostatic potential (ESP) mapping for the interaction between TMSP and F^- ions in the electrolyte.

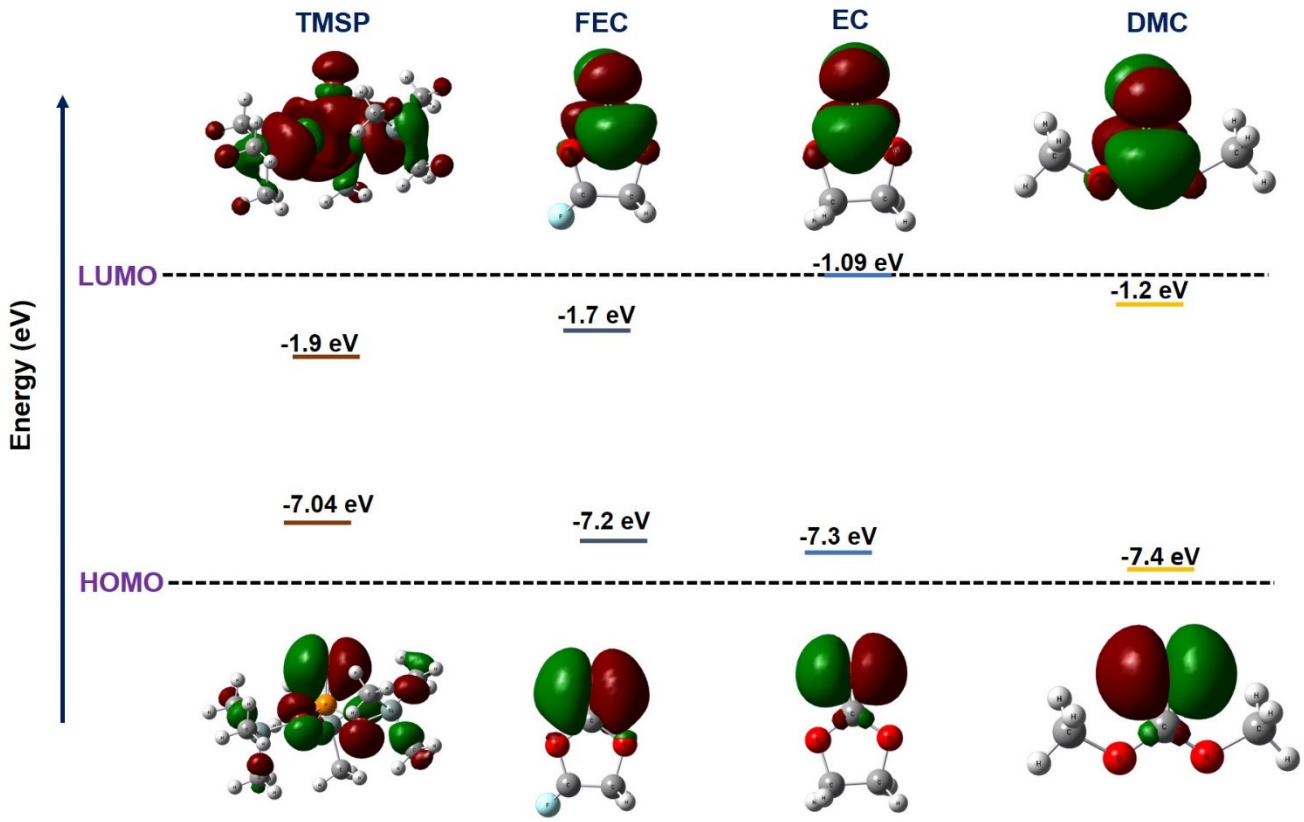


Figure S7 Shows the HOMO-LUMO energy levels of TMSP, FEC, EC, and DMC.

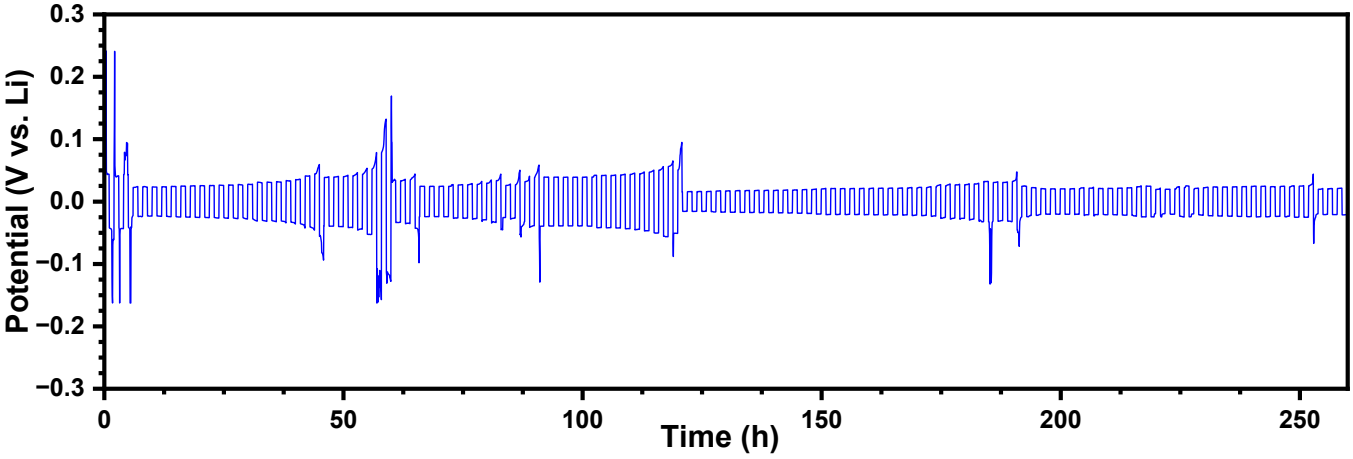


Figure S8 Symmetric cell study of Li/G-SPE_TMSP- 3 wt.%/Li at a current density of 1 mA cm^{-2} and capacity limit of 1 mAh cm^{-2}.

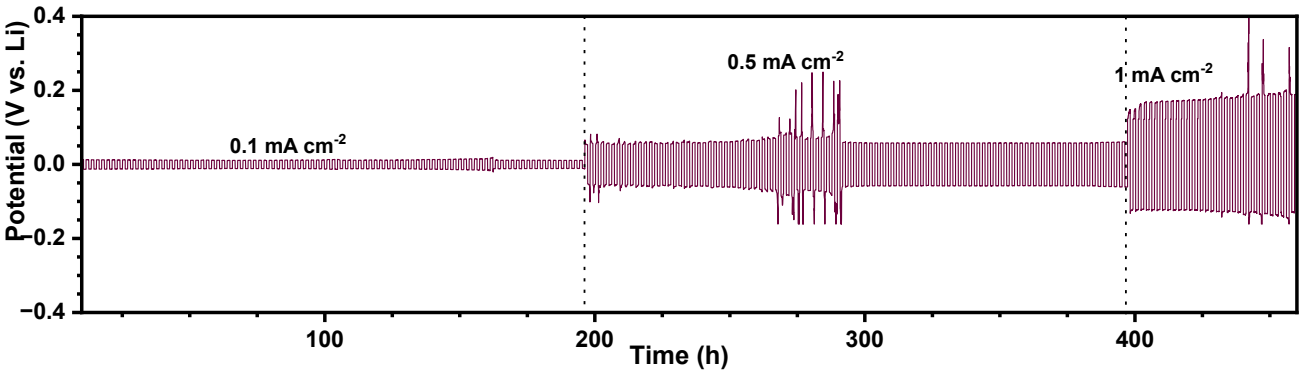


Figure S9 Symmetric cell study of Li/G-SPE_TMSP- 3 wt.%/Li at different arial current densities of 0.1, 0.5, 1 mA cm^{-2} with a capacity limit of 0.1, 0.5, and 1 mAh cm^{-2}.

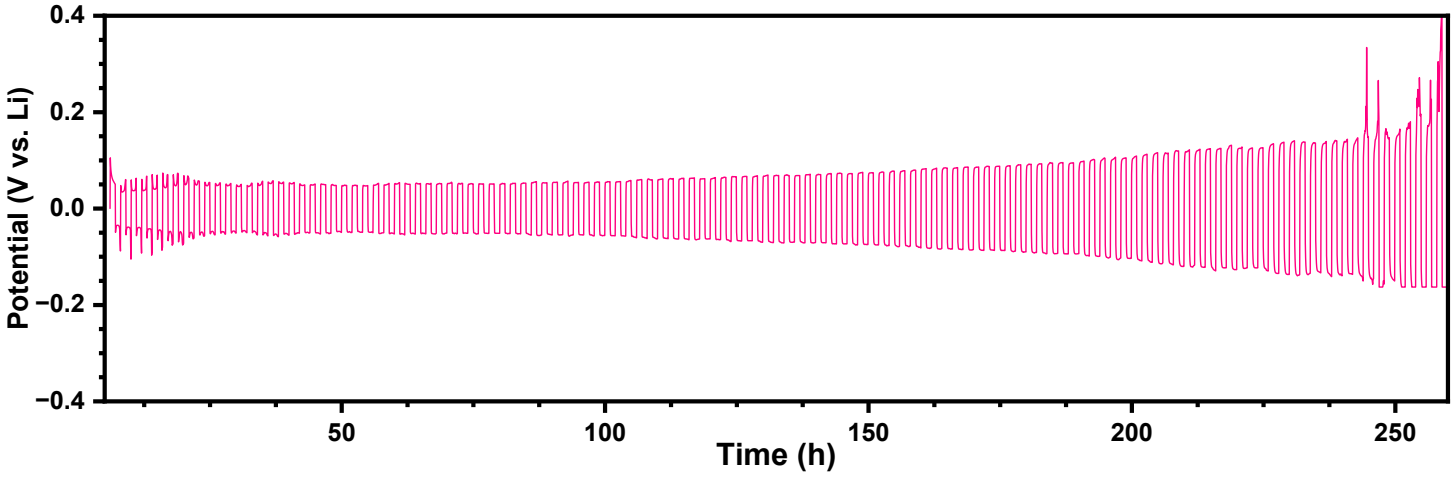


Figure S10 Asymmetric cell study with configuration Li/G-SPE_TMSP- 3 wt.%/Cu at a current density of 0.1 mA cm^{-2} with a capacity limit of 0.1 mAh cm^{-2}.

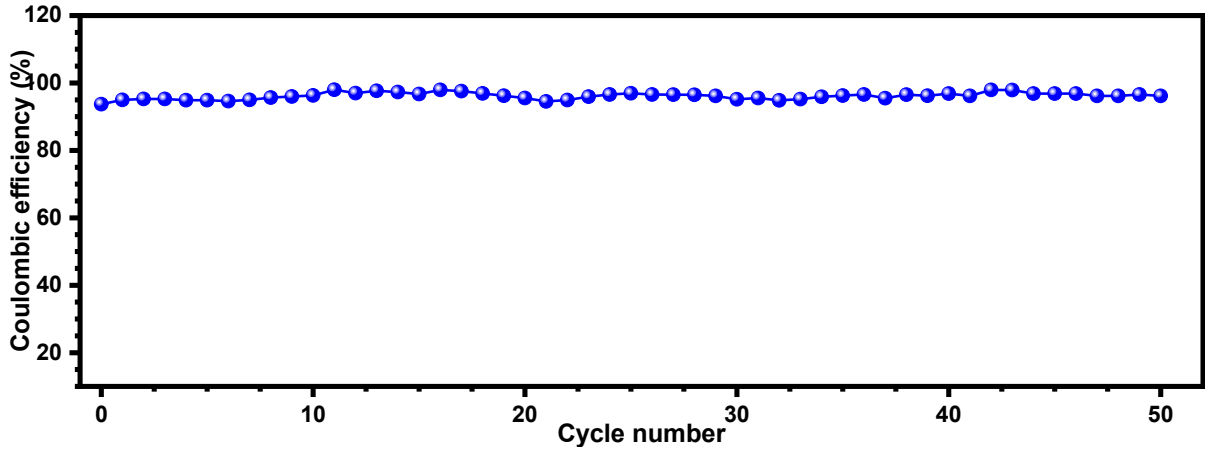


Figure S11 Coulombic efficiency *vs.* cycle number plot for the Li/Cu cell at a current density of 0.1 mA cm^{-2}.

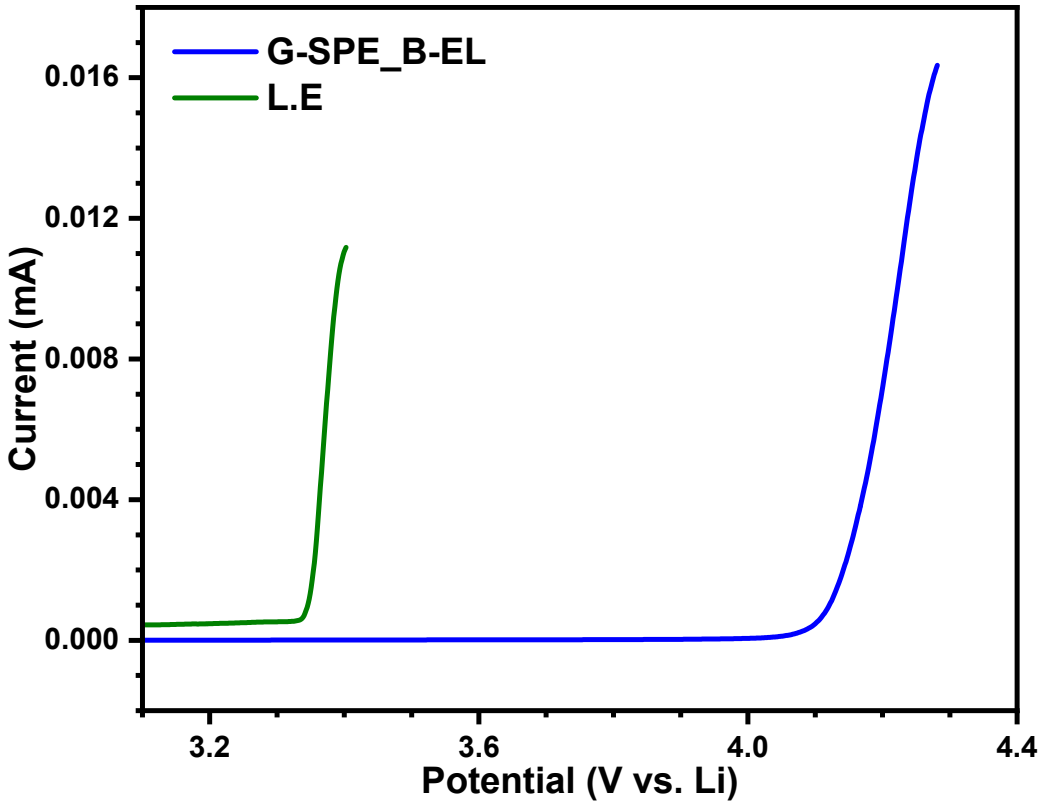


Figure S12 LSV plot for L.E and G-SPE_B-EL

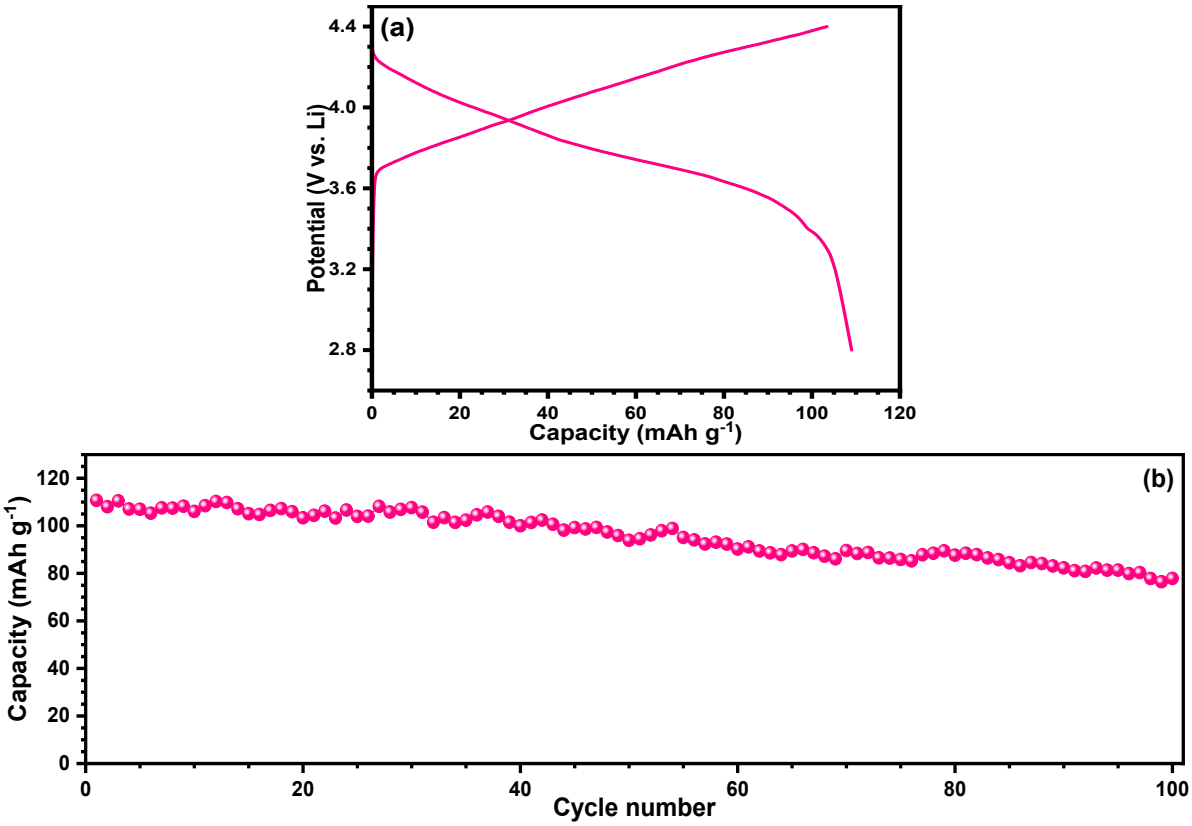


Figure S13 Plot showing the (a) potential *vs*. capacity and (b) capacity *vs*. cycle number of NMA cycled within the potential window of 2.8 – 4.4 V at a current density of 100 mA g^{-1}.

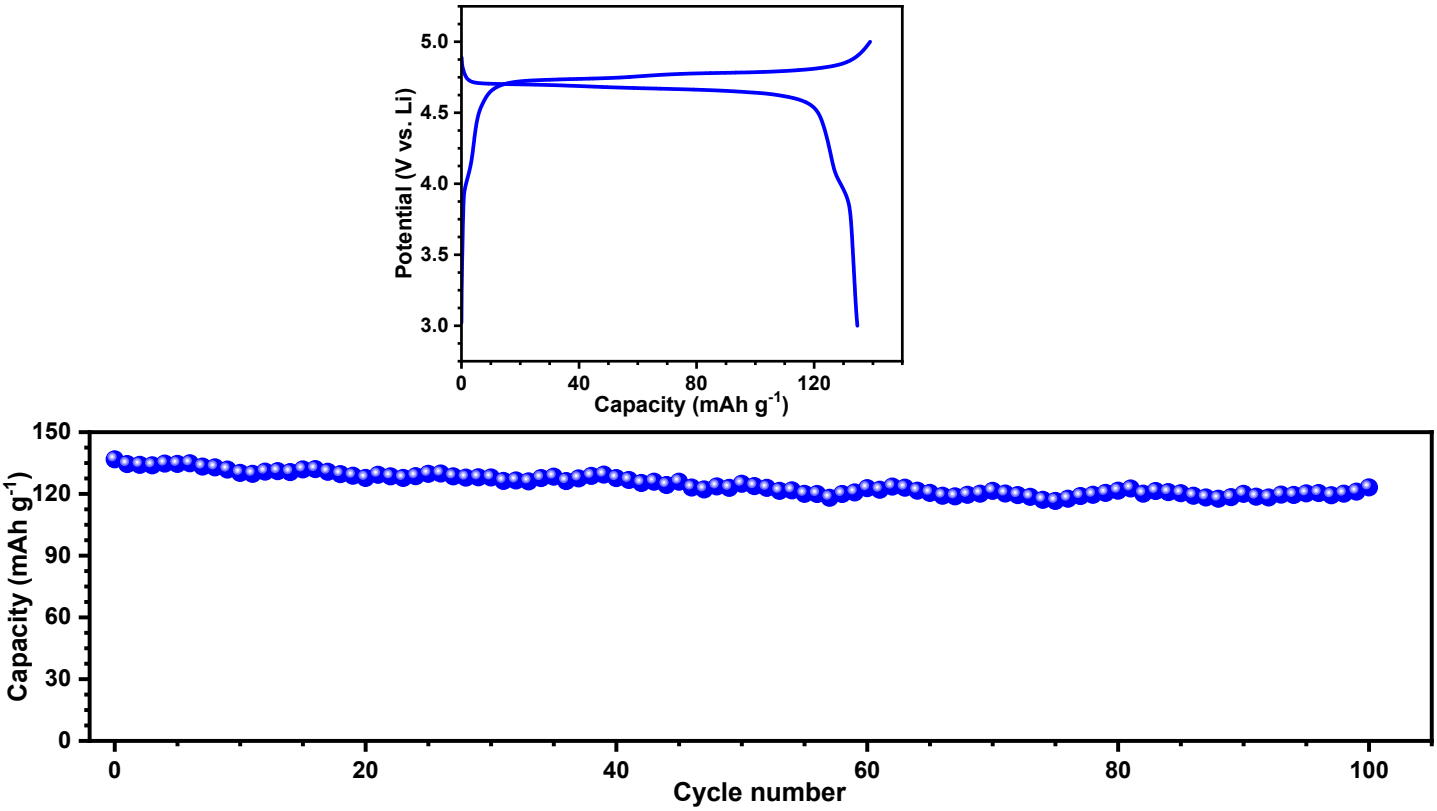


Figure S14 GCD profile of $LiNi_{0.5}Mn_{1.5}O_4$/Li using G-SPE_TMSP: 1.5 wt.% within a potential window of 3 – 5 V *vs*. Li at a current density of 50 mA g^{-1}.

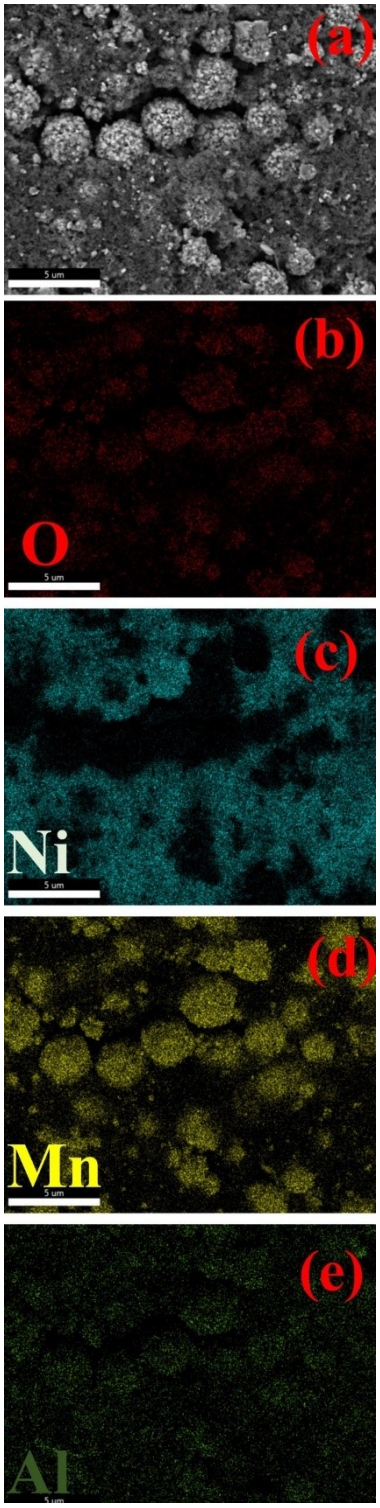


Figure S15 EDS analysis in cycled NMA electrodes for (a) electrode, (b) O, (c) Ni, (d) Mn, (e) Ni.

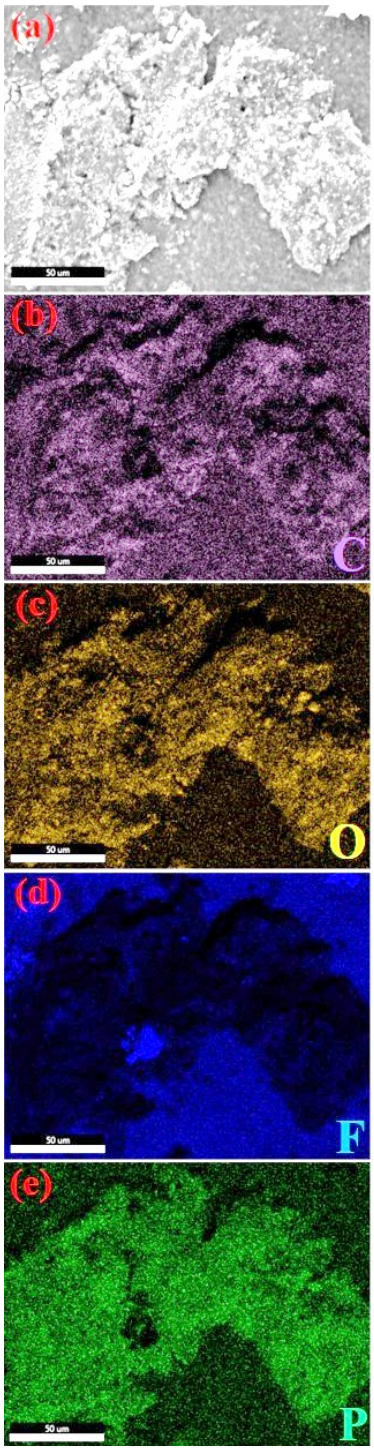


Figure S16 EDS elemental analysis of cycled G-SPE_TMSP for (a) cycled G-SPE_TMSP, (b) C, (c) O, (d) F, and (e) P.

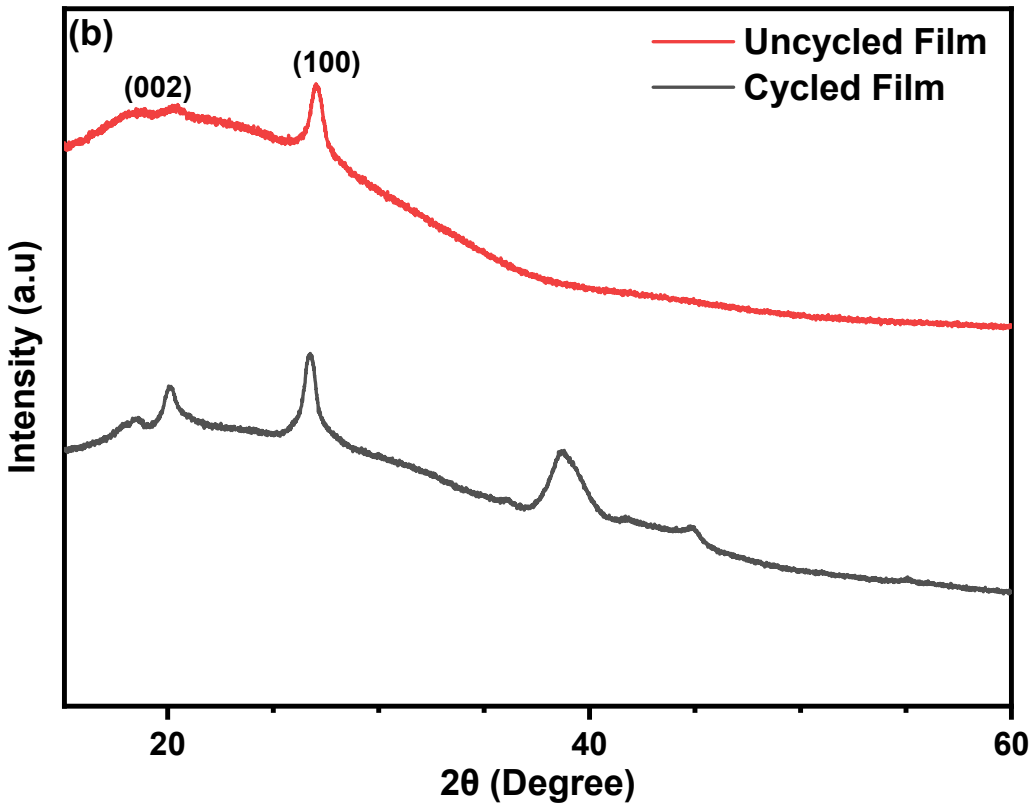


Figure S17 XRD analysis of cycled and uncycled G-SPE_TMSP- 3 wt.%.

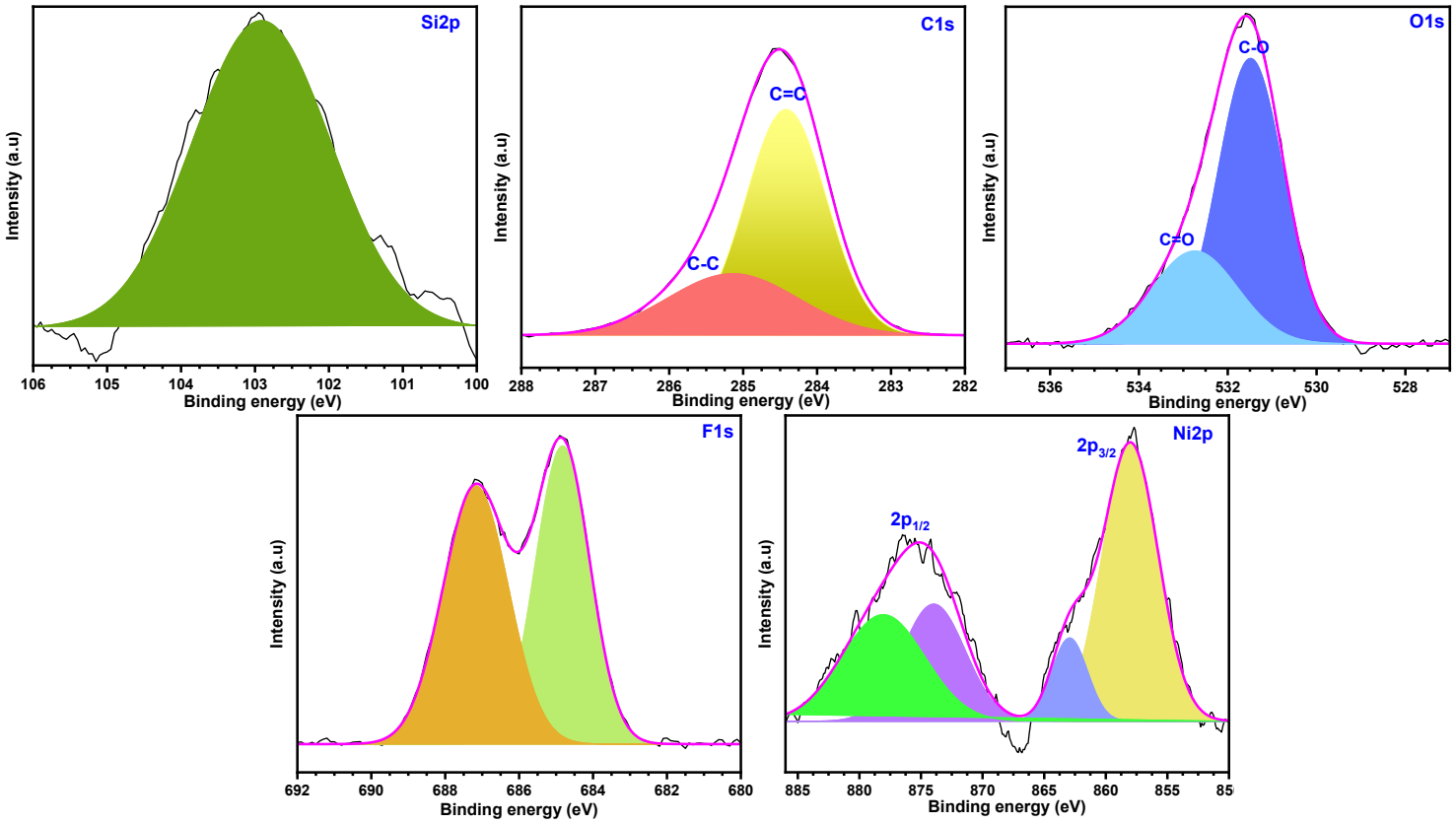


Figure S18 XPS analysis of the cycled NMA electrode.

Table T1 Shows the interaction between the TMSP additive and F^- ions in the electrolyte

Donor	Type	Donor Occupancy	Acceptor	Type	Acceptor Occupancy	E_2 (kcal mol^{-1})	$E_i - E_j$ (a.u)
F 45	LP (1)	0.993	P 1 — O 40	σ^*	0.105	10.41	0.19
F 45	LP (2)	0.979	P 1 — O 39	σ^*	0.102	6.07	0.35
F 45	LP (2)	0.979	C-27 — Si 42	σ^*	0.026	7.29	0.23
F 45	LP (3)	0.966	P 1 — O 39	σ^*	0.102	1.57	0.58
F 45	LP (3)	0.966	C 35 — Si 42	σ^*	0.024	3.49	0.39
F 45	LP (3)	0.966	O 41 — Si 42	σ^*	0.039	2.31	0.47
F 45	LP (3)	0.966	P 1 — O 2	σ^*	0.091	3.30	0.75
F 45	CR (1)	0.999	P 1 — O 40	σ^*	0.105	1.01	24.54

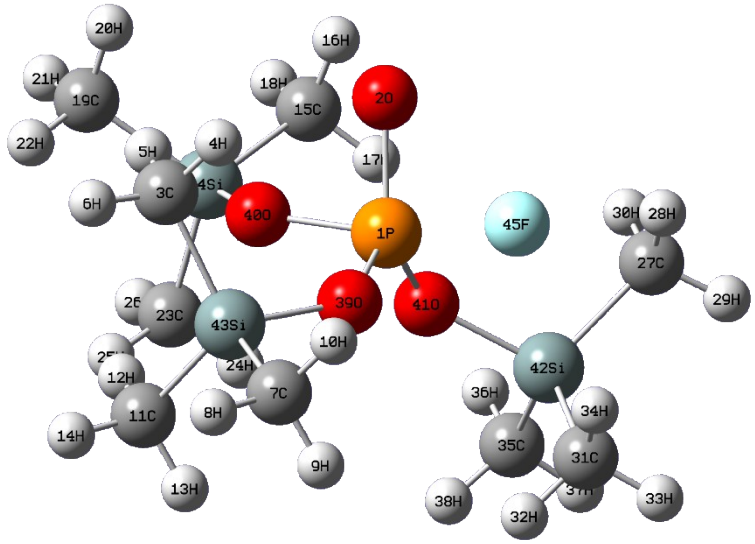


Table T2 % elemental composition of C, Al, Mn, and Ni in the uncycled and cycled NMA electrode.

Elements	Wt.% of elements (Uncycled electrode)	Wt.% of elements (Cycled electrode)
O	55.5	66.8
Al	0.5	0.3
Mn	1.4	0.9
Ni	5.5	3.4

Table T3 % elemental composition of C, O, F, and P in the uncycled and cycled G-

Elements	Wt.% of elements (Uncycled G-SPE_TMSP)	Wt.% of elements (Cycled G-SPE_TMSP)
C	52.4	53.3
O	1.3	16.3
F	40.5	22.8
P	0.1	4.3

SPE_TMSP- 3 wt.%.

Table T4 Fitting parameters of the EIS spectra of NMA/ G-SPE_TMSP(1-5 wt.%)/ Li for the 1st cycle corresponding to the equivalent circuit ($R_s + Q_1/R_{CT-1} + Q_2/R_{CT-2} + Z_W$).

G-SPE_TMSP	R_s	R_{CT-1}	Q_1	R_{CT-2}	Q_2	Z_W	X^2
1 wt.%	1.51	82.38	9.5×10^{-6}	17.4	1.3×10^{-3}	32.2	1.37
3 wt.%	8.22	96.55	8.8×10^{-6}	74.5	9.4×10^{-3}	45.35	1.07
5 wt.%	34.73	119.3	20.6×10^{-6}	118.4	0.3×10^{-3}	97.29	1.57

Table T5 Comparison based on ionic conductivity, transference number, and LSV data of the present report with previously reported works on gel-polymer electrolytes

Electrolyte (Polymer Filler \ Liquid electrolyte(L.E))	Ionic conductivity (mS cm^{-1})	Transference number	LSV (V)
This work	8.57	0.83	5.4
PAN_PEO \ 1M $LiPF_6$ in EC: DMC[37]	5.36	0.74	5
Cellulose _PVDF-HFP \ 1M $LiPF_6$ in EC: DMC[38]	1.89	0.89	5.35
PAN \ POSS \ 1M $LiPF_6$ in EC:DMC[39]	6.06	0.59	5.7
PVDF_MC \ 1M $LiPF_6$ in EC: DMC:EMC[40]	1.5	0.47	4.8
PAN-PMMA \ 1M $LiPF_6$ in EC: DMC[41]	5.1	0.5	5.2
PEGMA \ CTA[42]	1.8	0.7	5
Lignin fiber \ 1M $LiPF_6$ in EC: DMC: DEC[43]	3.73	0.85	7.2
Lignin _ PVP \ 1M $LiPF_6$ in EC: DMC: DEC[44]	2.52	0.56	5.3
CA_PLLA \ HNT \ 1M $LiPF_6$ in EC: DMC[45]	1.52	0.45	5.3
SPI_PVA [46]	3.8	0.32	5.5

Abbreviations:

CA: Cellulose acetate

PLLA: Poly-1-lactic acid \

HNT: Halloysite nanotube

PVA: Polyvinyl alcohol

SPI: Soy Protein Isolate

PVP: Polyvinyl pyrrolidone

PEGMA: Polyethylene glycol methacrylate

CTA: Cellulose triacetate

POSS: Polyhedral oligomeric silsesquioxane

PMMA: Polymethyl methacrylate

PAN: Polyacrylonitrile

PEO: Polyethylene oxide

MC: Methylcellulose