Supporting Information

A delicately-designed poly(vinylene carbonate-acrylonitrile) copolymer electrolyte enables 5 V lithium batteries

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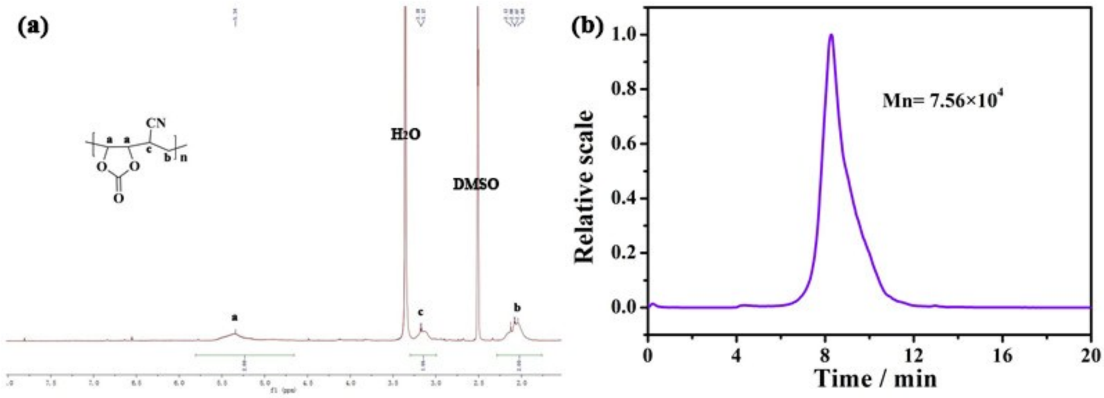

Fig. S1. a) ^{1}H NMR spectra and b) GPC of PVN.

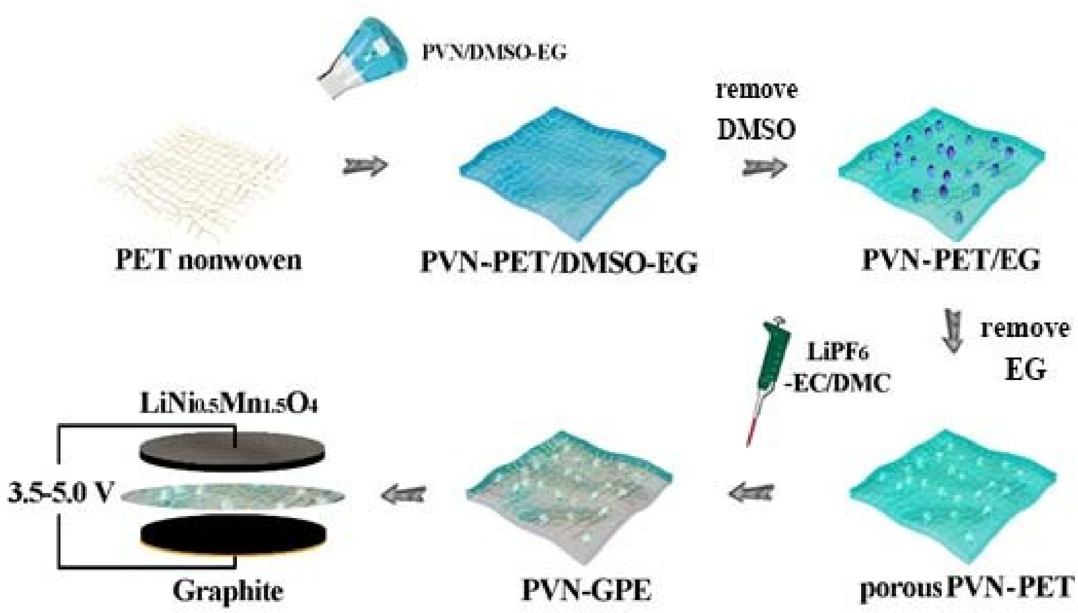

Fig. S2. The diagram of the preparation process for PVN-GPE.

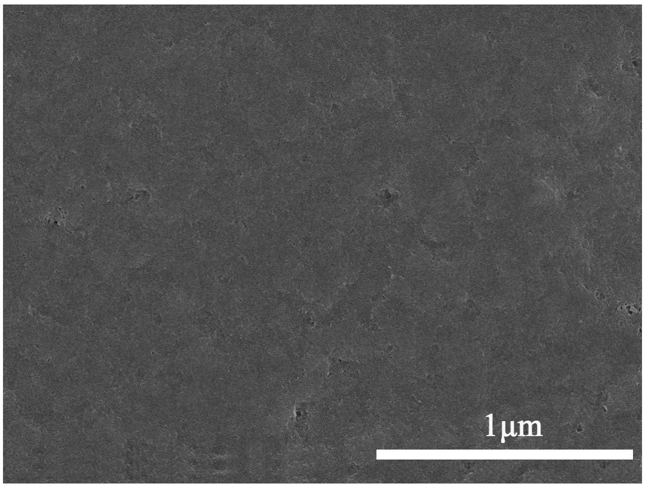

Fig. S3. The surface morphology of nonporous PVN-PET prepared without EG.

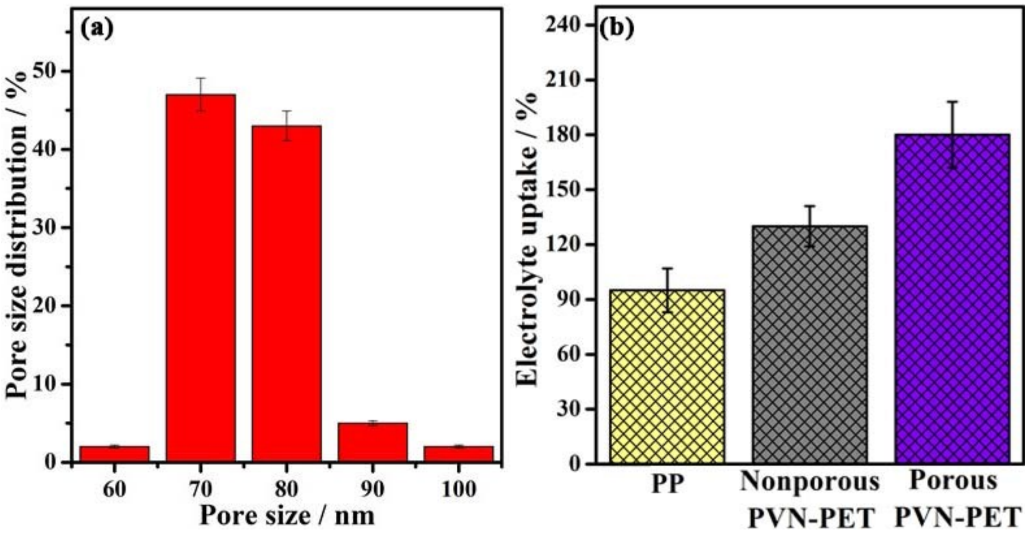

Fig. S4. a) The pore size distribution of porous PVN-PET and b) the electrolyte uptake of PP separator, porous and nonporous PVN-PET.

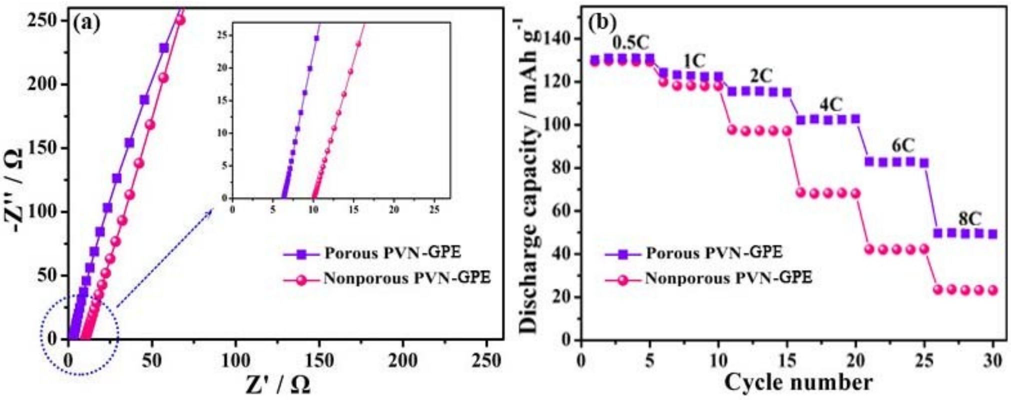

Fig. S5. a) AC impedance spectra comparison of porous and nonporous PVN-GPE; b) rate capability comparison of $LiNi_{0.5}Mn_{1.5}O_4$/Li cells using porous and nonporous PVN-GPE at 25 ^{o}C.

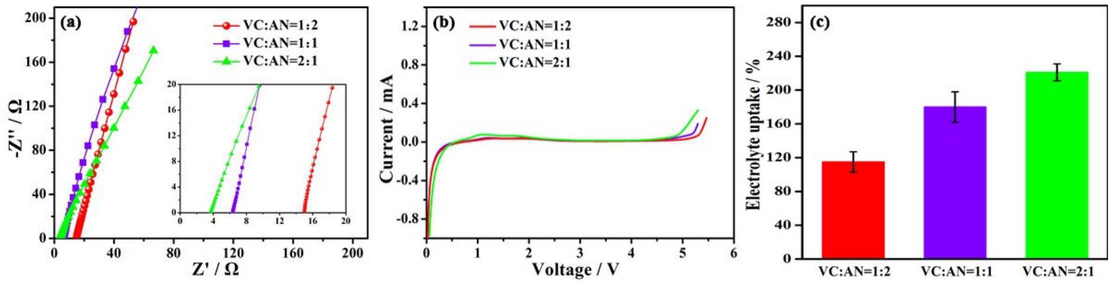

Fig. S6. a) AC impedance spectra comparison b) linear sweep voltammograms and c) the electrolyte uptake of PVN-GPE prepared with different molar ratios of VC to AN.

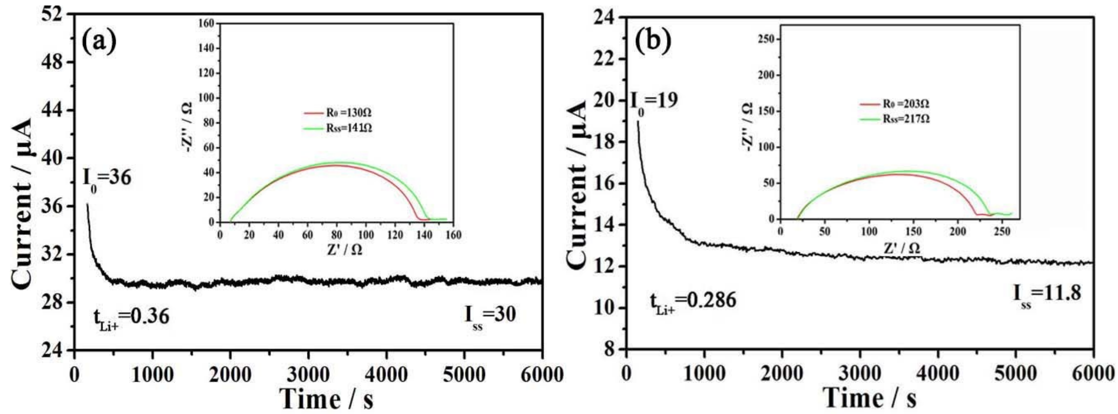

Fig. S7. Current-time curves following a DC polarization of 0.005 V for a) PVCA-GPE and b) PAN-GPE. Insets are Nyquist profiles of the electrochemical impedance spectroscopy response before and after polarization.

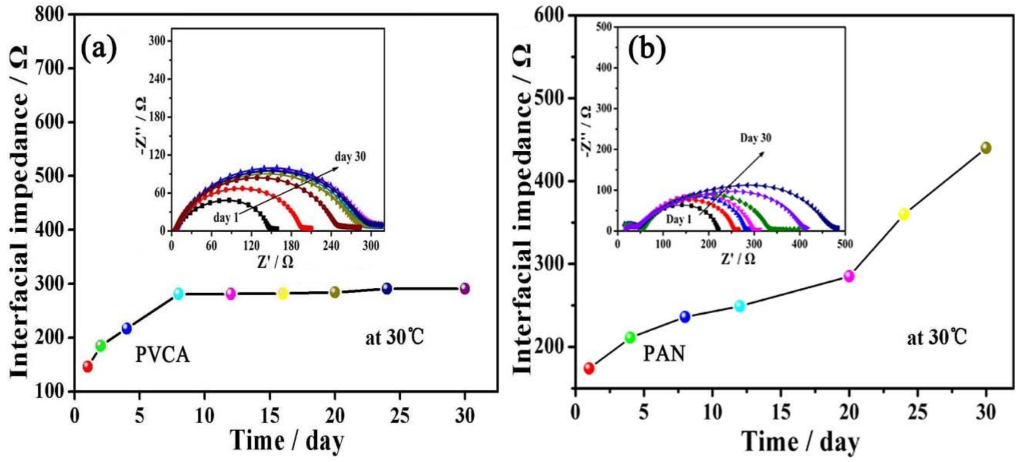

Fig. S8. Time evolution of the interfacial resistances within a month for a) PVCA-GPE and b) PAN-GPE. Insets are the corresponding EIS Nyquist plots.

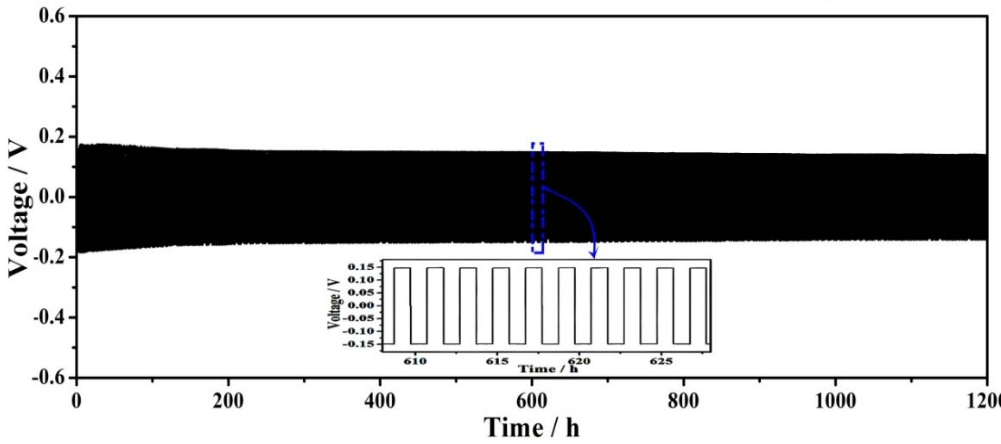

Fig. S9. The chronopotentiometry result of the Li/PVN-GPE/Li symmetric cell at room temperature with a current density of 2 mA cm^{-2} for 1 hr.

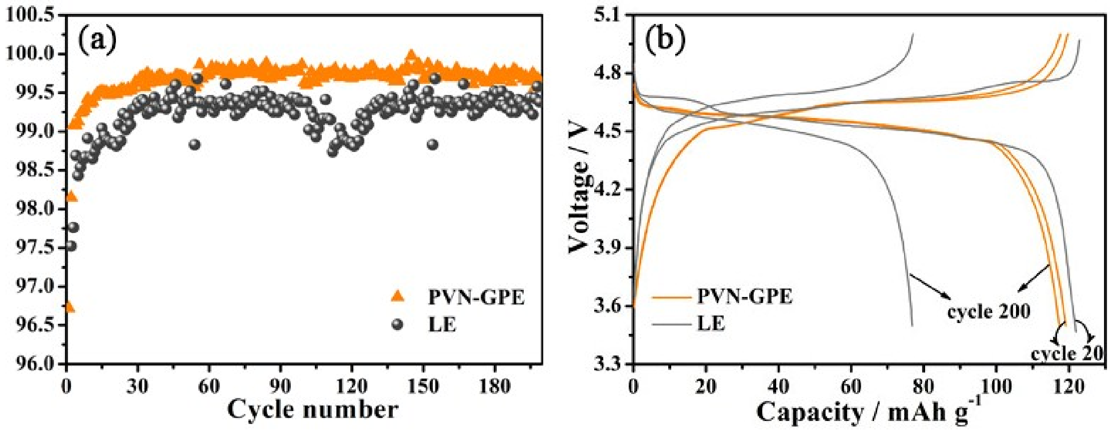

Fig. S10. a) Coulombic efficiencies and b) charge/ discharge curves of the 20th and 200th cycles for $LiNi_{0.5}Mn_{1.5}O_4$/graphite full cells assembled with PVN-GPE and LE.

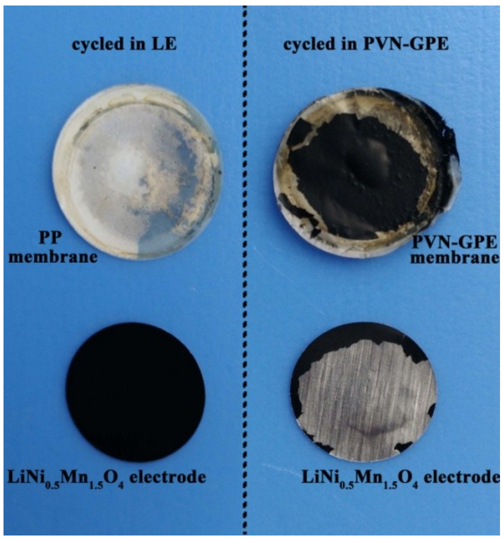

Fig. S11. Photographs of the surface of $LiNi_{0.5}Mn_{1.5}O_4$ electrodes and electrolyte membranes cycled in PVN-GPE and LE based cells after 200 cycles under 0.5 C.

The $LiNi_{0.5}Mn_{1.5}O_4$ electrodes and electrolyte membranes cycled in PVN-GPE and LE are carefully dismantled from the discharged full cells after 200 cycles under 0.5 C. The PP membrane cycled in LE is smoothly separated from the $LiNi_{0.5}Mn_{1.5}O_4$ electrode, whereas the PVN-GPE attaches tightly to the $LiNi_{0.5}Mn_{1.5}O_4$ electrode.

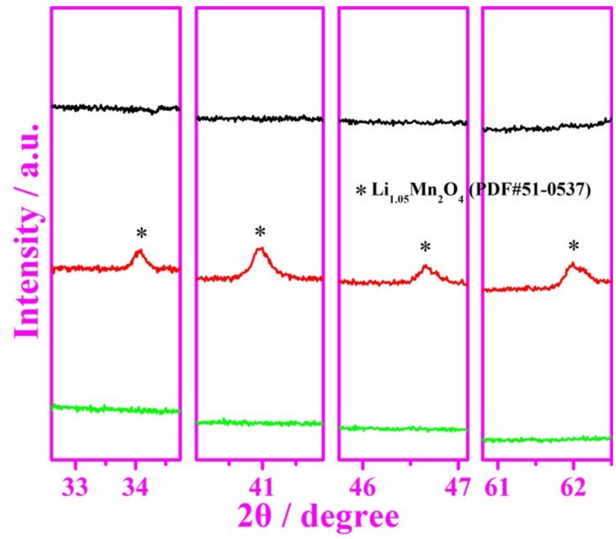

Fig. S12. Magnified views of selected regions in Fig. 5b. The black, red and green lines stand for pristine $LiNi_{0.5}Mn_{1.5}O_4$ cathodes, $LiNi_{0.5}Mn_{1.5}O_4$ cathodes cycled in LE and PVN-GPE based cells, respectively.

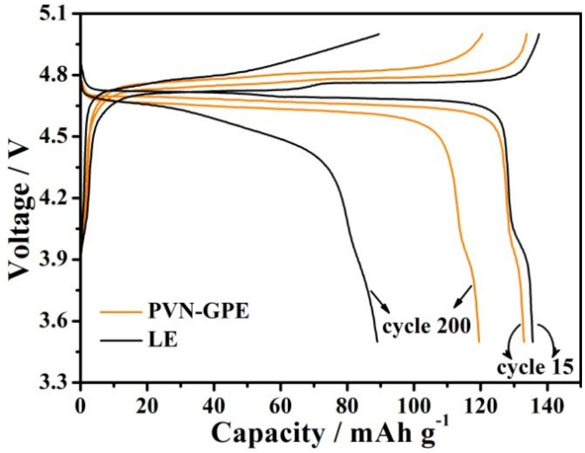

Fig. S13. Charge/discharge curves of the 15th and 200th cycles for $LiNi_{0.5}Mn_{1.5}O_4$/ Li batteries assembled with PVN-GPE and LE.

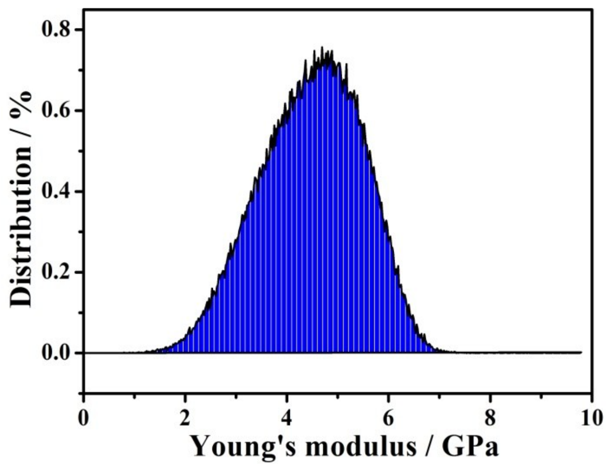

Fig. S14. The distribution of Young's modulus for PVN-GPE.

Table S1. The calculated values of ionic conductivity, decomposition potential and electrolyte uptake of PVN-GPE prepared with different molar ratios of VC to AN.

Molar ratio of VC to AN	Ionic conductivity / S cm^{-1}	Decomposition potential / V	Electrolyte uptake / %
1 : 2	1.32×10^{-4}	5.3	115
1 : 1	2.63×10^{-4}	5.2	180
2 : 1	5.4×10^{-4}	4.9	221

Table S2. The calculated values of VTF fitting parameters for PAN-GPE, PVCA-GPE and PVN-GPE.

	A/S·K^{½}·cm^{-1}	Ea/eV	T_0/K
PAN-GPE	0.0037	0.024	209
PVCA-GPE	0.00592	0.030	196
PVN-GPE	0.00611	0.021	206

Table S3 Fitting results of R_b, R_{SEI} and R_{ct} of $LiNi_{0.5}Mn_{1.5}O_4$/graphite full cells assembled with PVN-GPE and LE at the 20th and 200th cycles.

Elcetrolyte	Cycle	R_b/Ω	R_{SEI}/Ω	R_{ct}/Ω
PVN-GPE	20th	8.61	27.39	58.4
	200th	9.02	33.98	73.1
LE	20th	1.32	29.03	93.52

200^{th}	1.67	50.1	181.1

Table S4. The cyclability comparison of $LiNi_{0.5}Mn_{1.5}O_4$ based full cells with those of the representative reports.

Battery system (Voltage range)	Electrolyte	Cyclability (Retention cycles, rate)	Ref.
$LiNi_{0.5}Mn_{1.5}O_4$/graphite (3.5−5 V)	PVN-GPE	93.2% after 200 cycles, at 0.5 C	This work
$LiNi_{0.5}Mn_{1.5}O_4$/graphite (3.5−4.9 V)	poly(methylethyl α-cyanoacrylate) based GPE	91.5% after 100 cycles, at 0.5 C	17
$LiNi_{0.5}Mn_{1.5}O_4/Li_4Ti_5O_{12}$ (3.5−5 V)	PAMM based gel electrolyte	88.4% after 100 cycles, at 0.5 C	18
$LiNi_{0.5}Mn_{1.5}O_4$/graphite (3.8−5 V)	LiODFB:$LiBF_4$(4:1, by mol) /EC:EMC:DMC (1:1:1, by wt)	95.2% after 100 cycles, at 25 °C	36
$LiNi_{0.5}Mn_{1.5}O_4$/graphite (3.4−4.8 V)	1 wt% LiBOB+1 M $LiPF_6$/EC:DEC (1:2, by wt)	80% after 85 cycles, at 0.1 C	37
$LiNi_{0.5}Mn_{1.5}O_4$/graphite (3.5−4.9 V)	FEC-based electrolyte	~100% after 100 cycles, at C/3, at 55 °C	38