

# Crosslinked Gel Polymer Electrolyte from Trimethylolpropane Triglycidyl Ether by In Situ Polymerization for Lithium-Ion Batteries

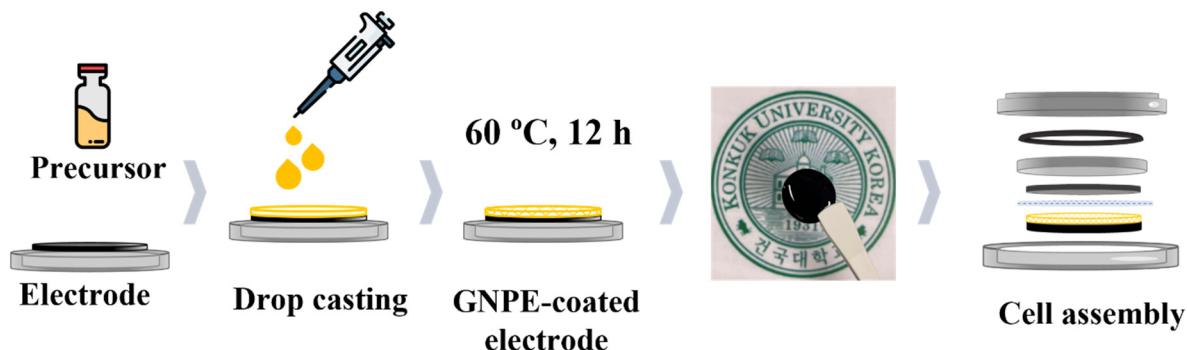


Figure S1. GNPEs preparation and cell assembly structure.

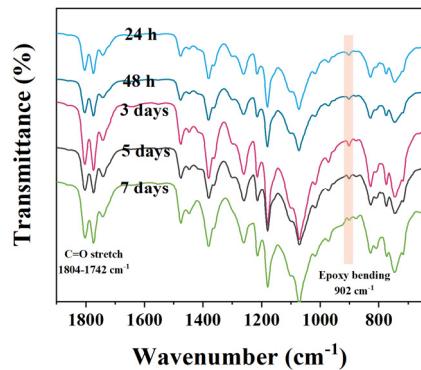
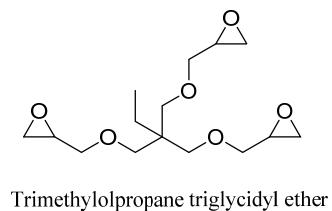


Figure S2. Chemical structure of TMPTGE and FT-IR resulting spectra of electrolyte with different times.

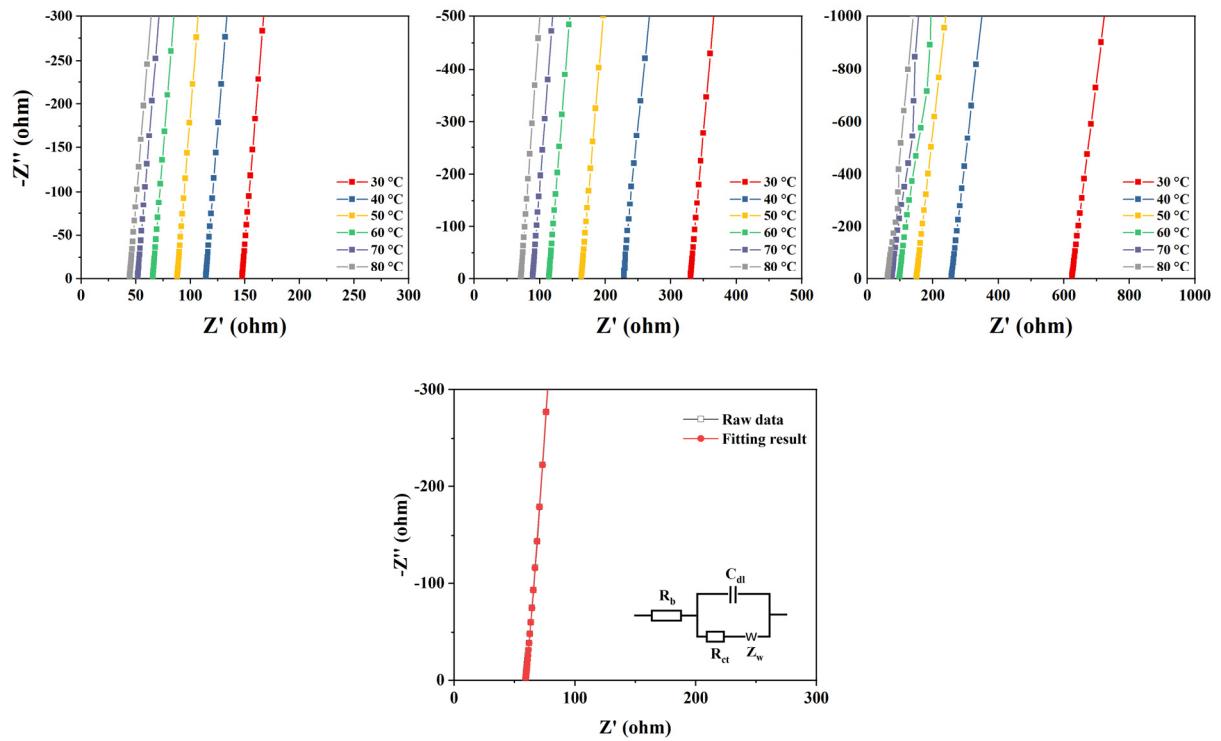


Figure S3. Nyquist plots of electrolytes at various temperatures and equivalent electric circuit model.

Table S1. Currents and resistances of the electrolytes for  $t_{Li^+}$  calculation.

Electrolyte	$I_s$ (A)	$I_0$ (A)	$\Delta V$ (mV)	$R_0$ (ohm)	$R_s$ (ohm)	$t_{Li^+}$
GNPE-1	2.18E-06	3.71E-06	0.1	145	213	0.58
GNPE-1.5	9.80E-07	2.11E-06	0.1	320	532	0.46
GNPE-2	1.03E-06	2.39E-06	0.1	640	897	0.42

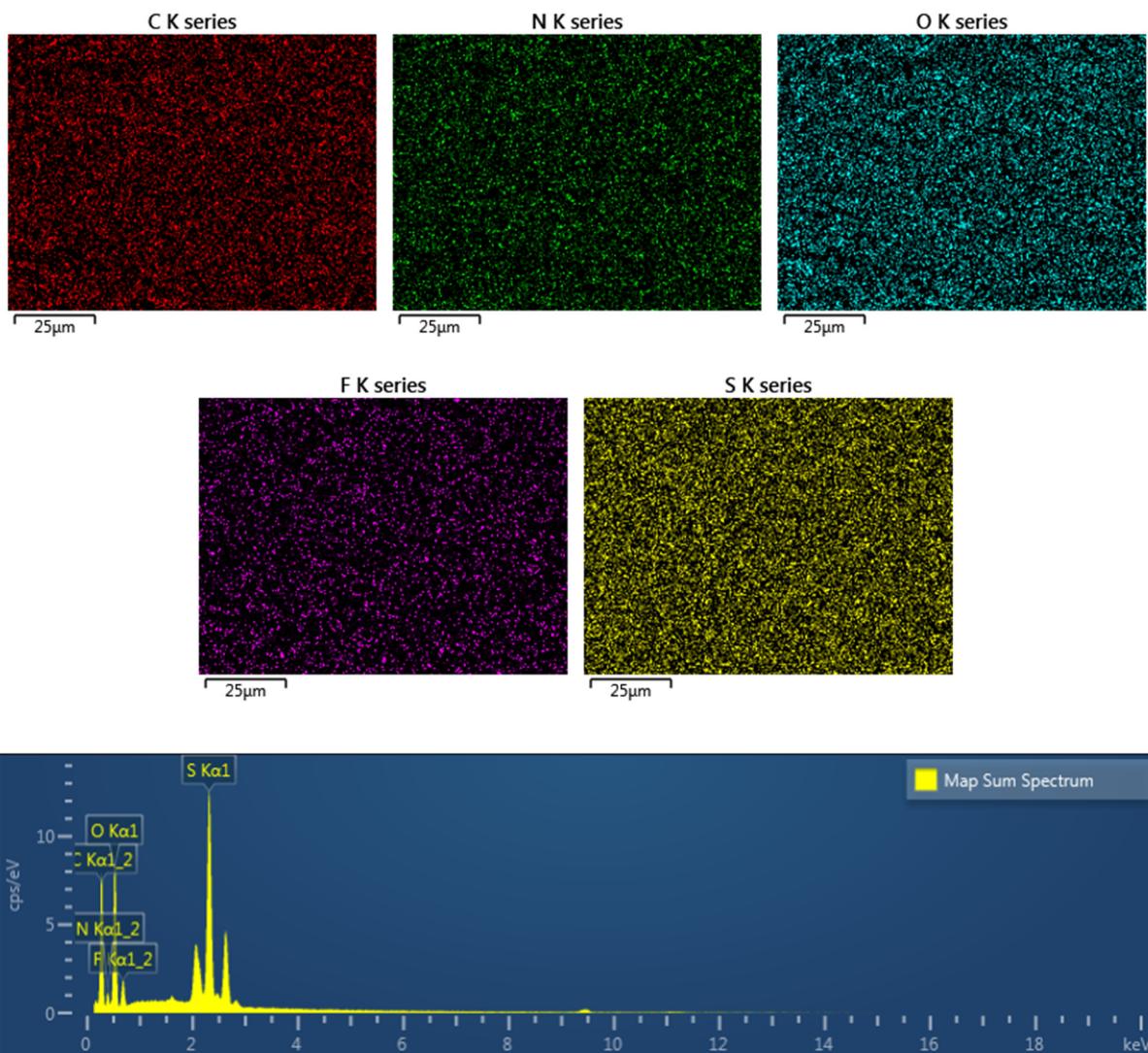
Bruce–Vincent–Evans Equation:

$$t_{Li^+} = I_s (\Delta V - I_0 R_0) / I_0 (\Delta V - I_s R_s)$$

where  $I_0$  and  $I_s$  are the initial and steady-state current, and  $R_0$  and  $R_s$  are denoted as the interfacial resistance between electrode and electrolyte of the symmetrical cell before and after polarization, respectively.

**Table S2.** Comparison of electrochemical properties of polymer electrolyte from reported works.

Polymer Electrolyte	Molecular Structure	$\sigma$ $S\ cm^{-1}$	$t^+$	Ref
<b>SPE</b>	PGO	$2.56 \times 10^{-8}$	0.549	[1]
	HPGO	$4.07 \times 10^{-7}$	0.693	
<b>In situ gel polymer</b>	PGTE	$4.16 \times 10^{-4}$	0.122	[2]
<b>In situ polymer</b>	PSEPE	$1.16 \times 10^{-4}$	0.61	[3]
<b>SPE</b>	PGA	$5.61 \times 10^{-4}$	0.43	[4]
<b>GPE</b>	DGEBA/L LTO	$2.02 \times 10^{-3}$	0.82	[5]
<b>GPE</b>	TiO <sub>2</sub> /Epoxy-based composite	$1.1 \times 10^{-4}$	0.661	[6]
<b>In situ gel polymer</b>	GNPE-1	$2.63 \times 10^{-4}$	0.58	This work
	GNPE-1.5	$1.17 \times 10^{-4}$	0.46	
	GNPE-2	$6.21 \times 10^{-5}$	0.42	



Element	Wt.%	Wt.% Sigma
C	42.71	0.54
N	11.3	0.7
O	30.37	0.43
F	5.37	0.21
S	10.24	0.14
Total:	100	

Figure S4. Elemental mapping and EDS analysis of GNPE-1.

## References

1. Yao, W.; Zhang, Q.; Qi, F.; Zhang, J.; Liu, K.; Li, J.; Chen, W.; Du, Y.; Jin, Y.; Liang, Y.; et al. Epoxy Containing Solid Polymer Electrolyte for Lithium Ion Battery. *Electrochim Acta* **2019**, *318*, 302–313, doi:10.1016/j.electacta.2019.06.069.
2. Ma, Y.; Sun, Q.; Wang, S.; ZPhou, Y.; Song, D.; Zhang, H.; Shi, X.; Zhang, L. Li Salt Initiated In-Situ Polymerized Solid Polymer Electrolyte: New Insights via in-Situ Electrochemical Impedance Spectroscopy. *Chemical Engineering Journal* **2022**, *429*, 132483, doi:10.1016/j.cej.2021.132483.
3. Jin, L.; Jang, G.; Lim, H.; Zhang, W.; Kim, W.; Jang, H. An In Situ Polymeric Electrolyte with Low Interfacial Resistance on Electrodes for Lithium-Ion Batteries. *Adv Mater Interfaces* **2022**, *9*, 1–11, doi:10.1002/admi.202101958.
4. Yu, T.Y.; Yeh, S.C.; Lee, J.Y.; Wu, N.L.; Jeng, R.J. Epoxy-Based Interlocking Membranes for All Solid-State Lithium Ion Batteries: The Effects of Amine Curing Agents on Electrochemical Properties. *Polymers (Basel)* **2021**, *13*, doi:10.3390/polym13193244.
5. Li, M.; Li, H.; Lan, J. Le; Yu, Y.; Du, Z.; Yang, X. Integrative Preparation of Mesoporous Epoxy Resin-Ceramic Composite Electrolytes with Multilayer Structure for Dendrite-Free Lithium Metal Batteries. *J Mater Chem A Mater* **2018**, *6*, 19094–19106, doi:10.1039/c8ta06341a.
6. Li, S.; Jiang, H.; Tang, T.; Nie, Y.; Zhang, Z.; Zhou, Q. Improved Electrochemical and Mechanical Performance of Epoxy-Based Electrolytes Doped with Mesoporous TiO<sub>2</sub>. *Mater Chem Phys* **2018**, *205*, 23–28, doi:10.1016/j.matchemphys.2017.10.075.