

## Supporting Information

### ***In situ fabrication of fluorine-modified acrylate-based gel polymer electrolytes for lithium-metal batteries***

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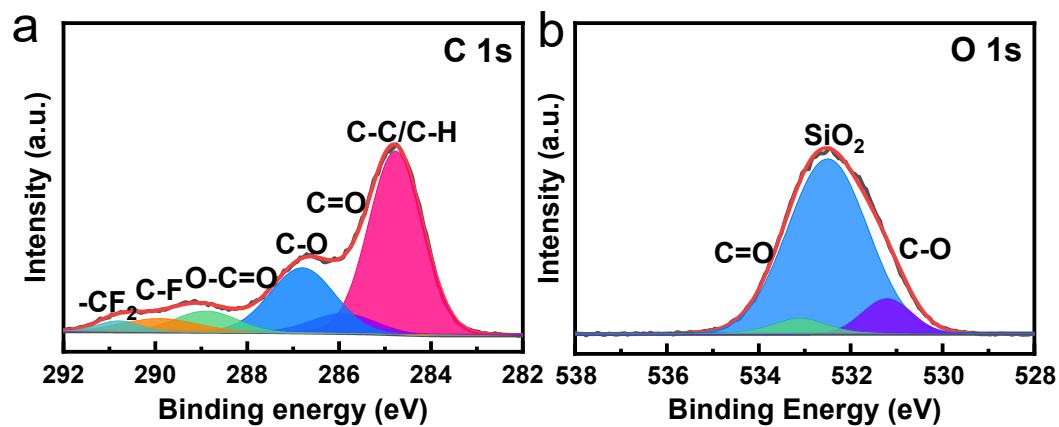
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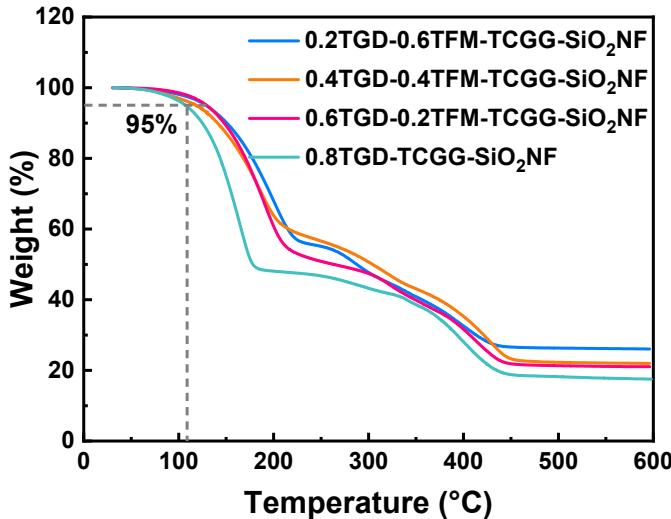
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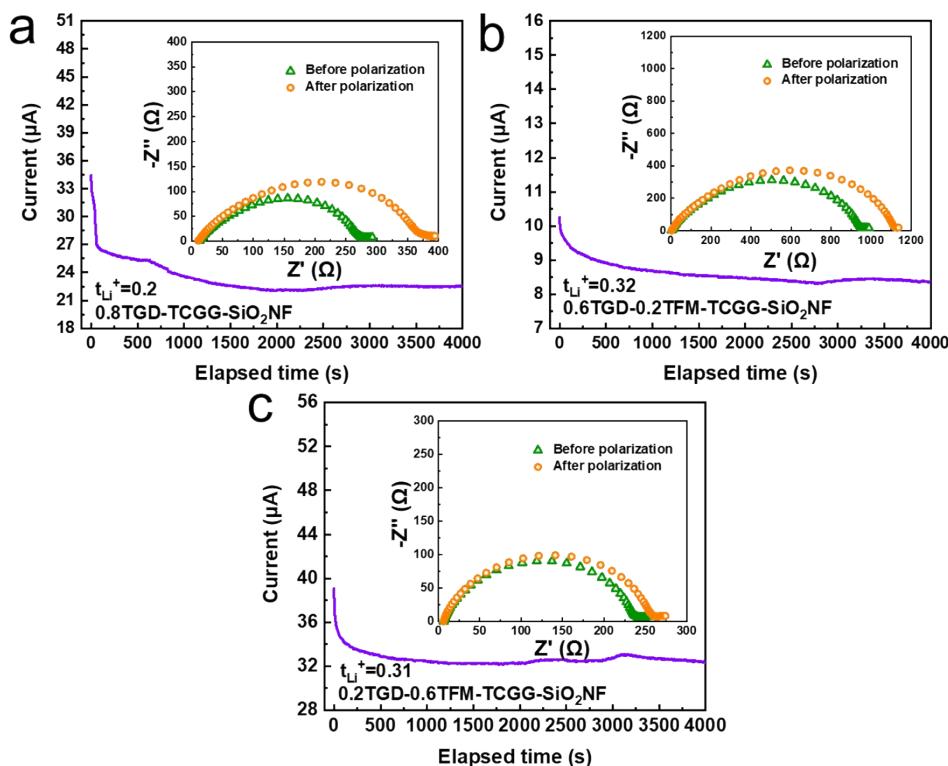
**Fig. S1.** Digital images of GPEs before and after thermal polymerization.



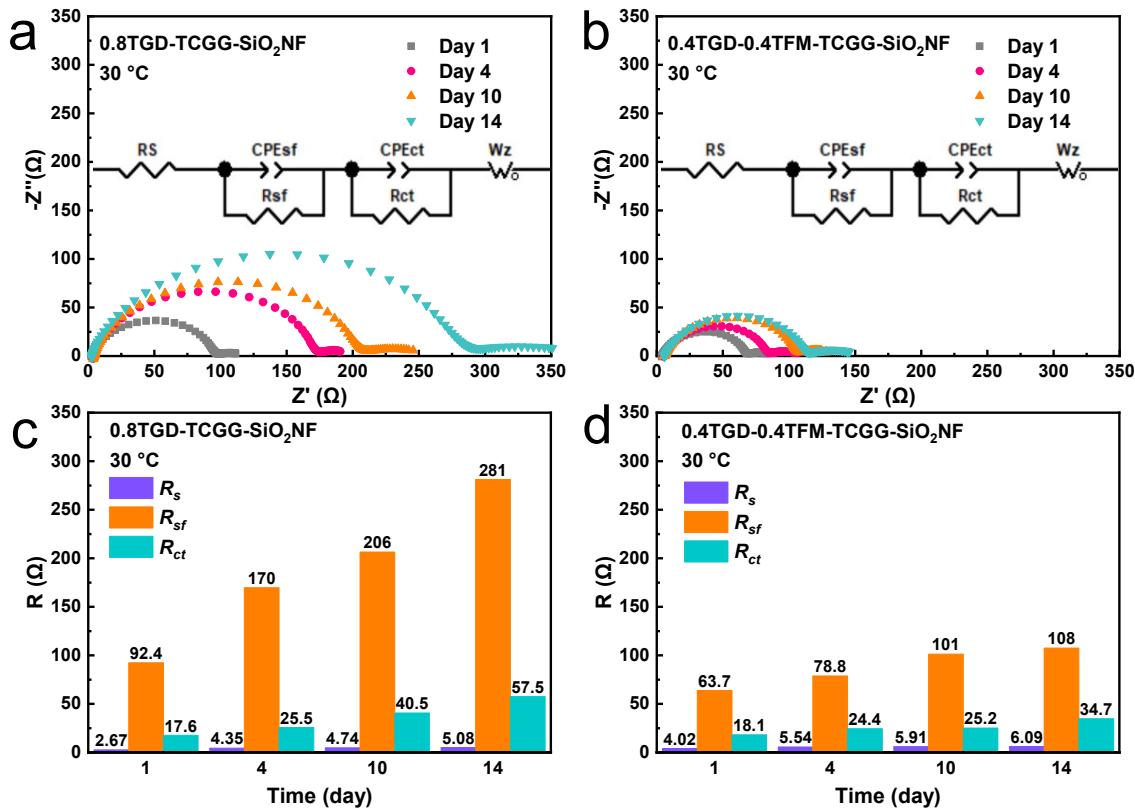
**Fig. S2.** XPS spectra on (a) C 1s and (b) O 1s of 0.4TGD-0.4TFM-TCGG-SiO<sub>2</sub>NF.



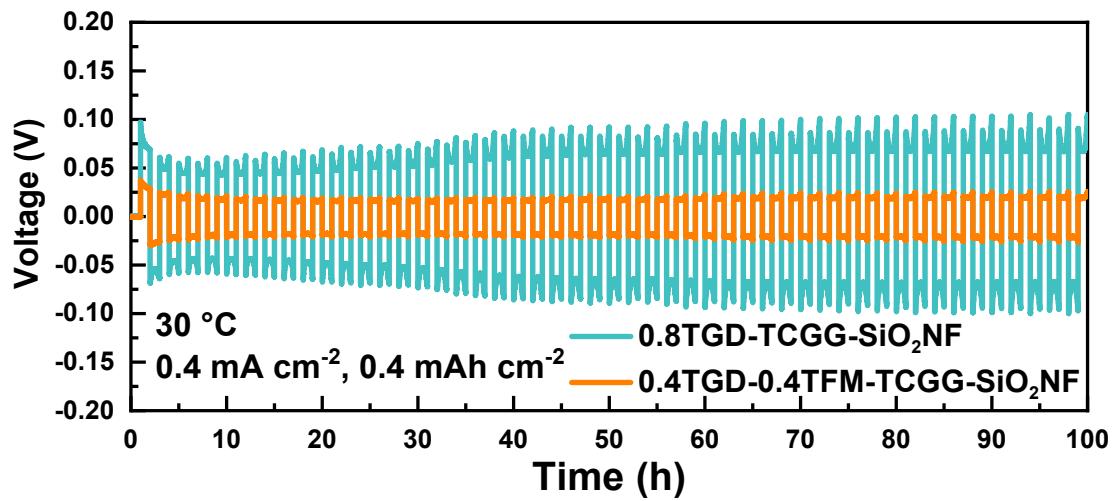
**Fig. S3.** TG curves of 0.8TGD-TCGG-SiO<sub>2</sub>NF, 0.6TGD-0.2TFM-TCGG-SiO<sub>2</sub>NF, 0.4TGD-0.4TFM-TCGG-SiO<sub>2</sub>NF, and 0.2TGD-0.6TFM-TCGG-SiO<sub>2</sub>NF.



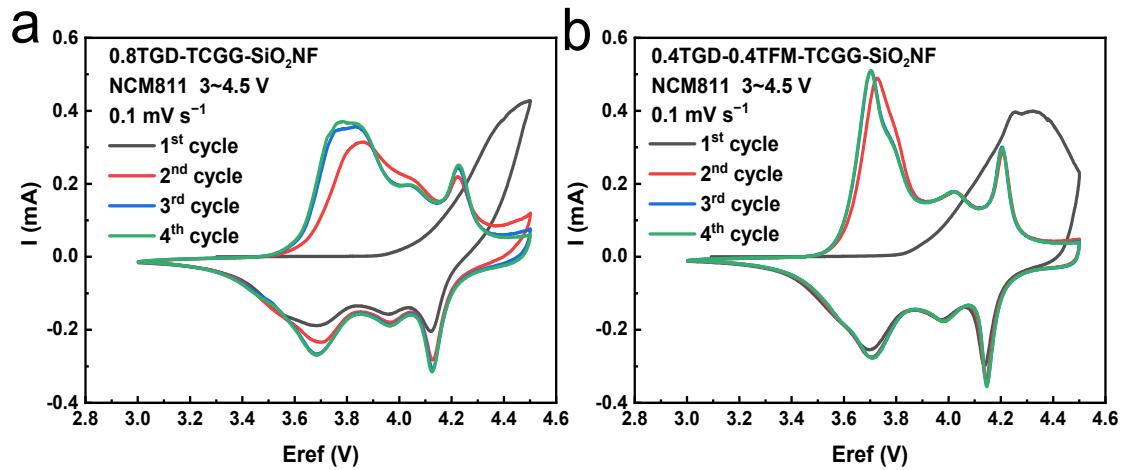
**Fig. S4.** Current and EIS curves of (a) 0.8TGD-TCGG-SiO<sub>2</sub>NF, (b) 0.6TGD-0.2TFM-TCGG-SiO<sub>2</sub>NF, and (c) 0.2TGD-0.6TFM-TCGG-SiO<sub>2</sub>NF before and after polarization at 25 °C.



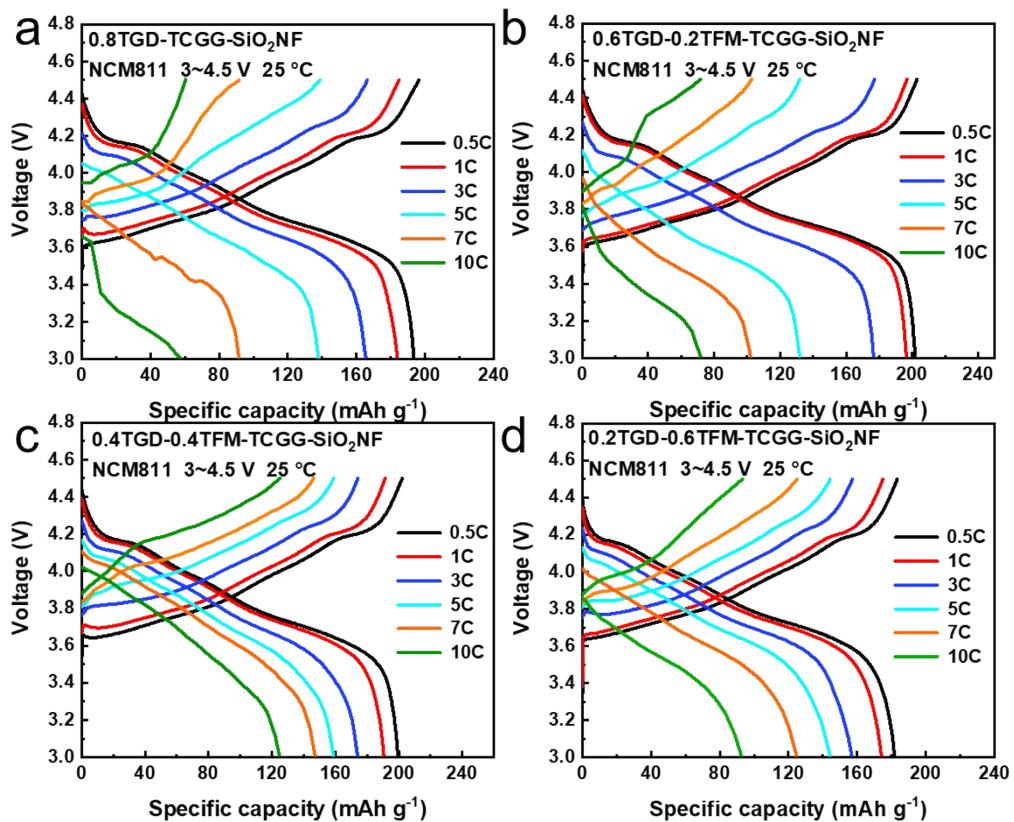
**Fig. S5.** EIS curves and fitting results of Li|0.8TGD-TCGG-SiO<sub>2</sub>NF|Li and Li|0.4TGD-0.4TFM-TCGG-SiO<sub>2</sub>NF|Li cells at 30 °C.



**Fig. S6.** Galvanostatic cycling profiles of Li|0.8TGD-TCGG-SiO<sub>2</sub>NF|Li and Li|0.4TGD-0.4TFM-TCGG-SiO<sub>2</sub>NF|Li cells at the current densities of 0.4 mA cm<sup>-2</sup> with a restricted specific capacity of 0.4 mAh cm<sup>-2</sup> at 30 °C.

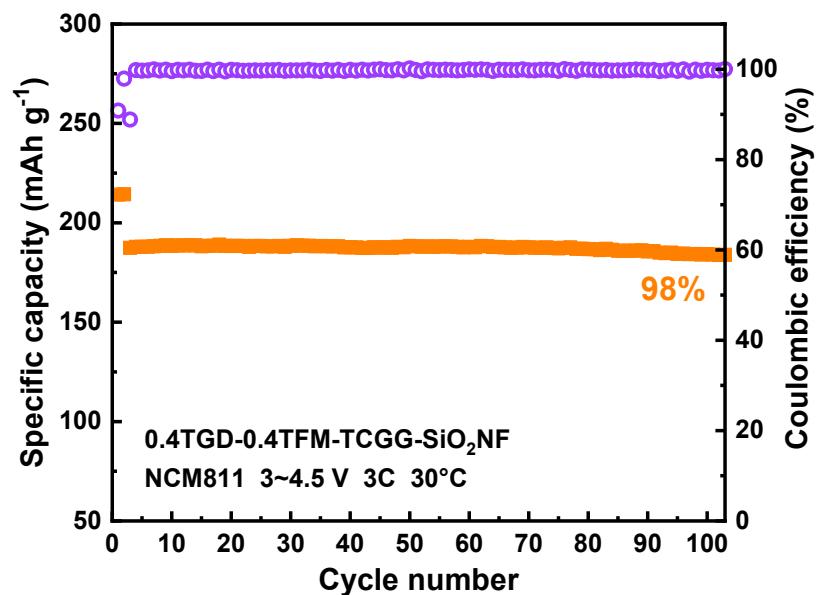


**Fig. S7.** CV curves of (a) NCM811|0.8TGD-TCGG-SiO<sub>2</sub>NF|Li and (b) NCM811|0.4TGD-0.4TFM-TCGG-SiO<sub>2</sub>NF|Li batteries at 3.0 ~ 4.5 V with a scan rate of 0.1 mV s<sup>-1</sup> at 30 °C.

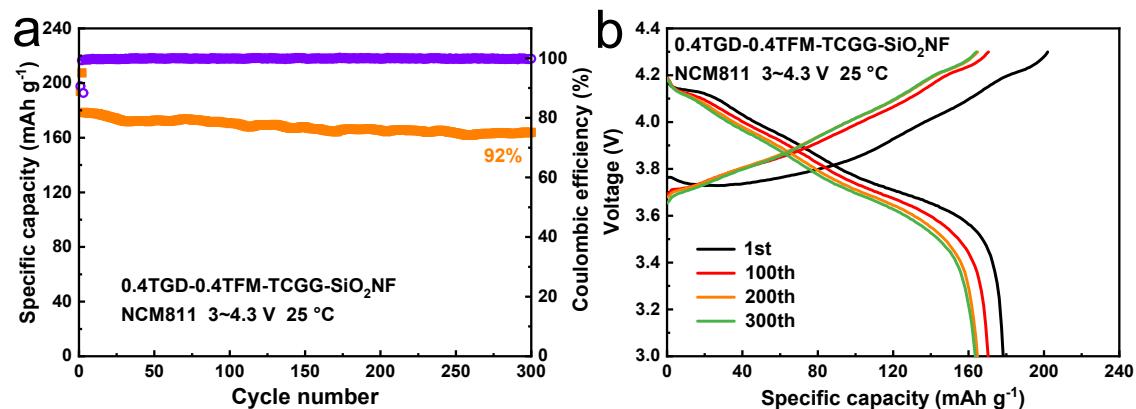


**Fig. S8.** Voltage profiles of (a) NCM811|0.8TGD-TCGG-SiO<sub>2</sub>NF|Li, (b) NCM811|0.6TGD-0.2TFM-TCGG-SiO<sub>2</sub>NF|Li, (c) NCM811|0.4TGD-0.4TFM-

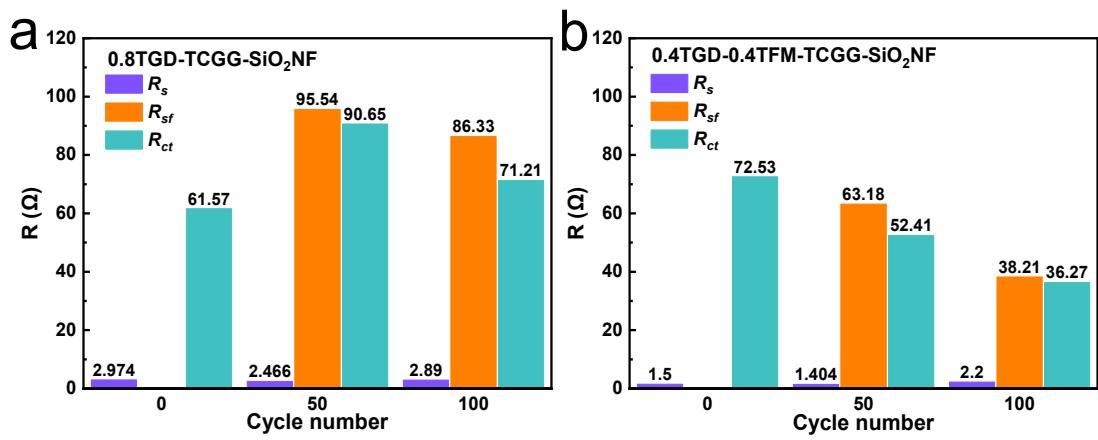
TCGG-SiO<sub>2</sub>NF|Li, and (d) NCM811|0.2TGD-0.6TFM-TCGG-SiO<sub>2</sub>NF|Li batteries at various C-rate.



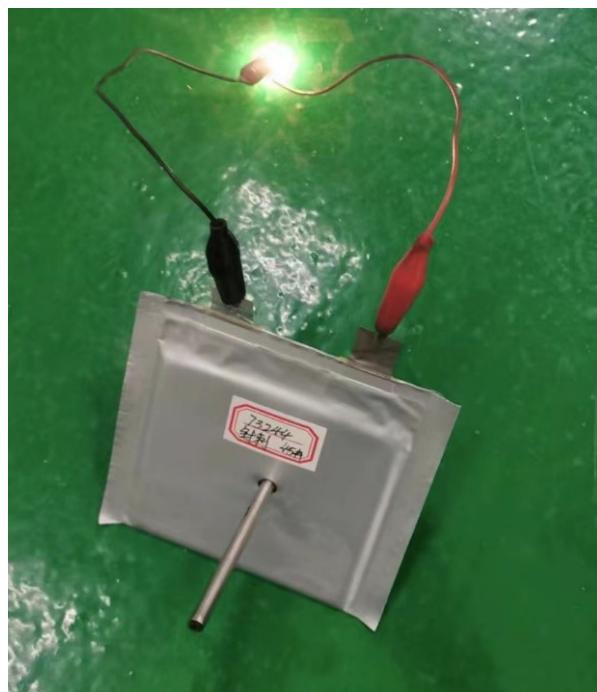
**Fig. S9.** Cycling performance of NCM811|0.4TGD-0.4TFM-TCGG-SiO<sub>2</sub>NF|Li battery at 3C.



**Fig. S10.** (a) Cycling performance and (b) voltage profiles of NCM811|0.4TGD-0.4TFM-TCGG-SiO<sub>2</sub>NF|Li battery at 2C.



**Fig. S11.**  $R_s$ ,  $R_{sf}$ , and  $R_{ct}$  variation of NCM811|0.8TGD-TCGG-SiO<sub>2</sub>NF|Li and NCM811|0.4TGD-0.4TFM-TCGG-SiO<sub>2</sub>NF|Li batteries.



**Fig. S12.** Pinprick testing of pouch battery.

**Table. S1.** Composition of different GPEs.

	TGD (g)	TFM (g)	TCGG (g)	AIBN (mg)
0.8TGD-TCGG	0.8	0	4.5	8
0.6TGD-0.2TFM-TCGG	0.6	0.2	4.5	8
0.4TGD-0.4TFM-TCGG	0.4	0.4	4.5	8
0.2TGD-0.6TFM-TCGG	0.2	0.6	4.5	8

**Table. S2.** Ionic conductivities ( $\sigma$ ) of GPEs.

	0.8TGD-TCGG-SiO <sub>2</sub> NF (S cm <sup>-1</sup> )	0.6TGD-0.2TFM-TCGG-SiO <sub>2</sub> NF (S cm <sup>-1</sup> )	0.4TGD-0.4TFM-TCGG-SiO <sub>2</sub> NF (S cm <sup>-1</sup> )	0.2TGD-0.6TFM-TCGG-SiO <sub>2</sub> NF (S cm <sup>-1</sup> )
30 °C	1.67×10 <sup>-4</sup>	2.60×10 <sup>-4</sup>	3.56×10 <sup>-4</sup>	2.15×10 <sup>-4</sup>
40 °C	1.97×10 <sup>-4</sup>	2.74×10 <sup>-4</sup>	4.21×10 <sup>-4</sup>	2.57×10 <sup>-4</sup>
50 °C	2.35×10 <sup>-4</sup>	3.02×10 <sup>-4</sup>	4.70×10 <sup>-4</sup>	3.24×10 <sup>-4</sup>
60 °C	2.73×10 <sup>-4</sup>	3.10×10 <sup>-4</sup>	4.80×10 <sup>-4</sup>	3.58×10 <sup>-4</sup>
70 °C	3.05×10 <sup>-4</sup>	3.29×10 <sup>-4</sup>	5.00×10 <sup>-4</sup>	3.79×10 <sup>-4</sup>
80 °C	3.17×10 <sup>-4</sup>	3.53×10 <sup>-4</sup>	5.10×10 <sup>-4</sup>	4.10×10 <sup>-4</sup>

**Table. S3.** The electrochemical performances of this work and reported GPEs.

Battery	$t_{\text{Li}^+}$	Test conditions	Initial capacity (mAh g <sup>-1</sup> )	Cycles capacity retention	Reference
NCM811 TFEMA-VEC Li	0.44	3~4.5 V 25 °C	1C, 218	300 <sup>th</sup> , 70%	[1]
LFP PEGDMA-ETFP/Li	0.42	2.5~3.8 V 25 °C	0.5C, 142.1	100 <sup>th</sup> , 98%	[2]
NCA MA-HFMA-MMA Li	0.47	3~4.3 V 25 °C	0.5C, 162.2	200 <sup>th</sup> , 91%	[3]
LFP PEGDA-ETPTA Li	0.7	2.5~4.0 V 25 °C	1C, 124.4	200 <sup>th</sup> , 76%	[4]
LCO PVCA Li	0.57	2.5~4.3 V 50 °C	0.1C, 146.0	150 <sup>th</sup> , 82%	[5]
NCM811 0.4TGD-0.4TFM- TCGG-SiO <sub>2</sub> NF Li	0.57	3~4.3 V 25 °C	2C, 178.4	300 <sup>th</sup> , 92%	This work
		3~4.5 V 25 °C	2C, 184.2	260 <sup>th</sup> , 91%	

## References

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