

## **Supporting Information**

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Concentrated electrolytes widen the operating temperature range of lithium-ion batteries

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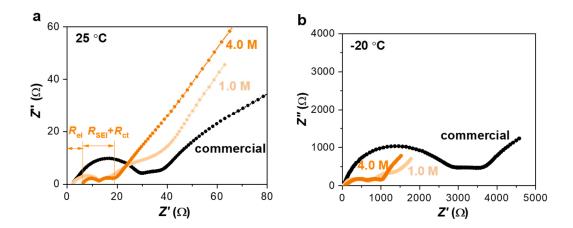
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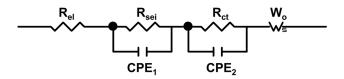
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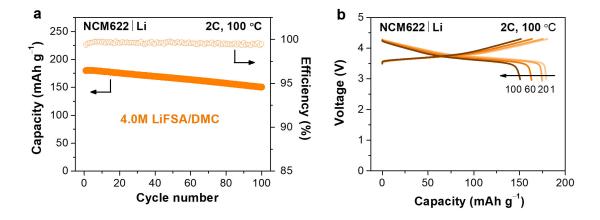


**Figure S1.** Impedance spectra of the graphite Li half-cells with various electrolytes at 25 °C (a) and -20 °C (b). The *x*-axis intercept is attributed to the electrolyte bulk resistance ( $R_{\rm el}$ ). The two semicircles are composed of the solid–electrolyte interphase (SEI) resistance ( $R_{\rm SEI}$ ) and charge-transfer resistance ( $R_{\rm ct}$ ). At 25 °C, the cell with 4.0 M LiFSA/dimethyl carbonate (DMC) shows larger  $R_{\rm el}$  but much smaller  $R_{\rm SEI}$  +  $R_{\rm ct}$  than those with 1.0 M LiFSA/DMC and the commercial electrolyte, leading to the lowest total cell resistance. At -20 °C, the total cell resistance is primarily dominated by  $R_{\rm SEI}$  and  $R_{\rm ct}$  because  $R_{\rm el}$  becomes negligible compared to  $R_{\rm SEI}$  and  $R_{\rm ct}$ .

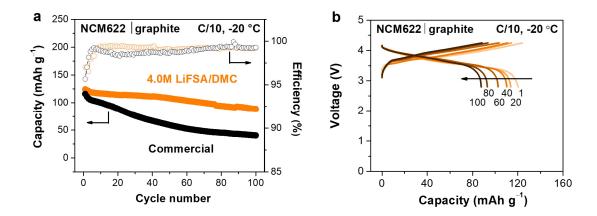


**Table S1.** The electrolyte bulk resistance ( $R_{el}$ ), solid–electrolyte interphase resistance ( $R_{SEI}$ ) and charge-transfer resistance ( $R_{ct}$ ) obtained from equivalent circuit fitting.

Electrolyte	Temperature (°C)	$R_{ m el}\left(\Omega ight)$	$R_{\mathrm{SEI}}\left(\Omega\right)$	$R_{\mathrm{ct}}\left(\Omega\right)$
1.0 M LiPF <sub>6</sub> /EC:DMC	-20	12.6	3872	2494
	25	3.0	22.8	156.4
1.0 M LiFSA/DMC	-20	30.9	543.5	3544
	25	2.2	9.4	262.1
4.0 M LiFSA/DMC	-20	77.2	431.7	701.8
	25	6.2	4.1	51.0



**Figure S2.** High-temperature operation of a NCM622|Li half-cell. (a) Cycling performance of the half-cell with 4.0 M LiFSA/DMC at 2C and 100 °C. (b) Selected charge—discharge curves for the half-cell.



**Figure S3.** Low-temperature operation of a NCM622|graphite full cell. (a) Cycling performance of the full cell with 4.0 M LiFSA/DMC and commercial 1.0 M LiPF<sub>6</sub>/EC:DMC at C/10 and - 20 °C. (b) Selected charge–discharge curves for the full cell with 4.0 M LiFSA/DMC at -20 °C.

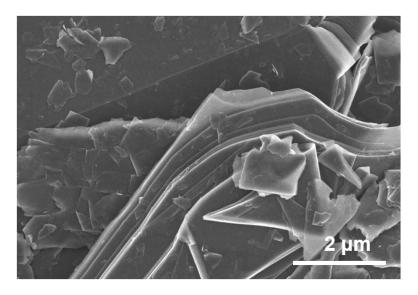


Figure S4. SEM images of the as-prepared graphite electrode.

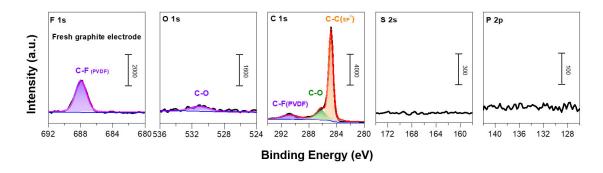


Figure S5. X-ray photoelectron spectra of the as-prepared graphite electrode.

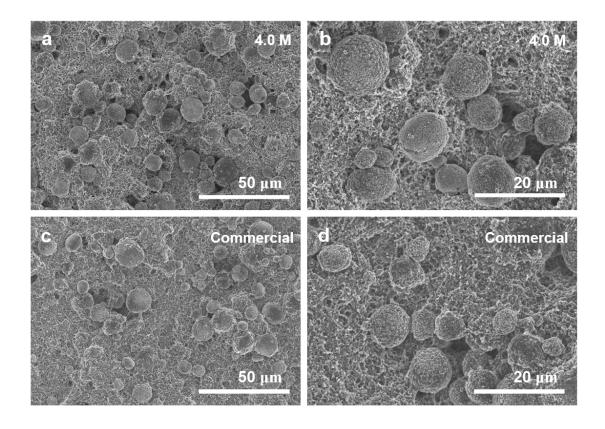
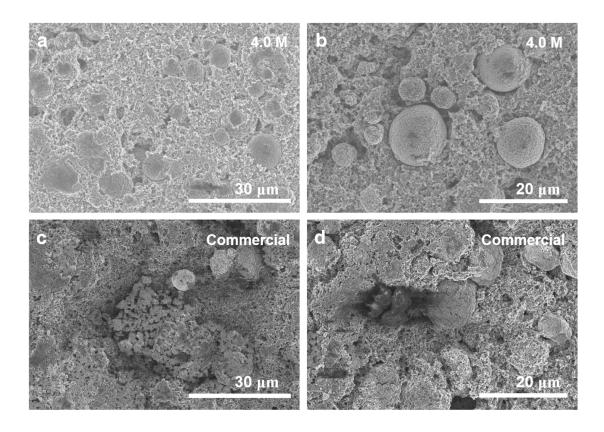


Figure S6. SEM images of the NCM622 electrodes after cycling at 25 °C. (a, b) are SEM images of NCM622 electrodes cycled in the concentrated 4.0 M LiFSA/DMC electrolyte. (c, d) are SEM images of NCM622 electrodes cycled in the commercial 1.0 M LiFF6/EC:DMC (1:1) electrolyte.



**Figure S7. SEM images of the NCM622 electrodes after cycling at 100 °C. (a, b)** are SEM images of NCM622 electrodes cycled in the concentrated 4.0 M LiFSA/DMC electrolyte. **(c, d)** are SEM images of NCM622 electrodes cycled in the commercial 1.0 M LiPF<sub>6</sub>/EC:DMC (1:1) electrolyte.

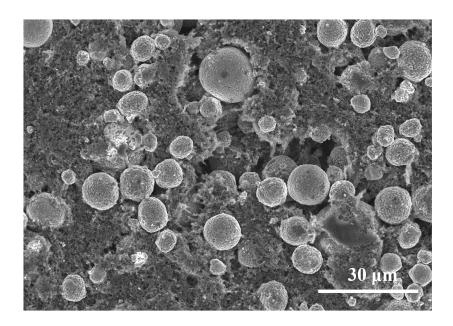


Figure S8. SEM images of the as-prepared NCM622 electrode.