Supporting Information

Synergistic sulfonyl-ether polyimide binders for stabilized high-loading NCM811 cathodes in high-energy lithium-ion batteries

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Supplementary Note S1

EIS method: According to the Warburg diffusion in the low-frequency region, the diffusion coefficient of the Li^+ ion(D) is calculated using the following equation:

$$D = 0.5\left(\frac{RT}{n^2F^2ACA_w}\right)^2 \quad (1)$$

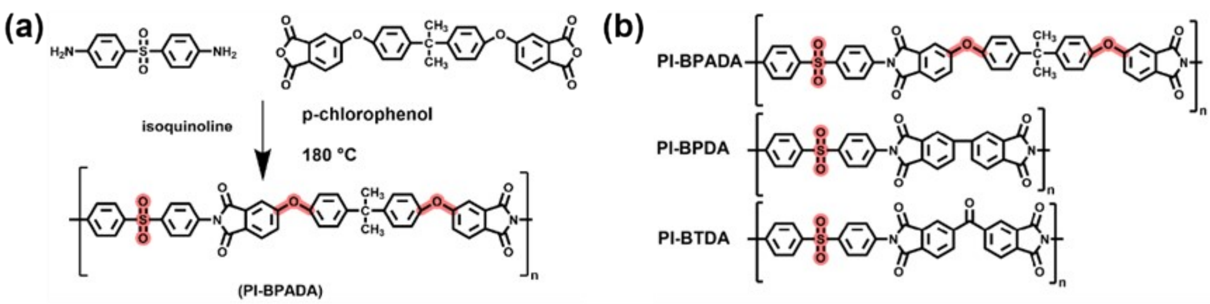
where R is the ideal gas constant, T is the absolute temperature, F is the Faraday constant, C is the concentration of Li^+ in the unit cell, A is the surface area of electrode in cm^2, n is the number of electrons per molecule during the charge/discharge process, and Aw is the Warburg factor, which is related to Z'

Supplementary Note S2

GITT method: The lithium diffusion coefficient (D_{Li^+}) for the cathodes is derived from Equation 2:

$$D_{Li^+} = \frac{4}{\pi\tau}\left(\frac{m_BV_m}{M_BS}\right)^2\left(\frac{\Delta E_s}{\Delta E_\tau}\right)^2 \quad (2)$$

Where τ(s) is the duration of the current pulse. m_B (g) and M_B (g mol^{-1}) represent the mass and molecular weight for the material, respectively.V_m(cm^3 mol-^{1}) is the molar volume for the active material, obtained from standard crystallographic data. S (cm^2) is the surface area of the active material, obtained from standard crystallographic data.S (cm^2) is the surface area of the active material in the cathode. ΔE_s is the steady-state voltage change due to the current pulse, and ΔE_τ is the voltage change during the constant current pulse.



Scheme S1. (a) PI-BPADA binder synthesis. (b) Chemical structures of synthesized PI binders.

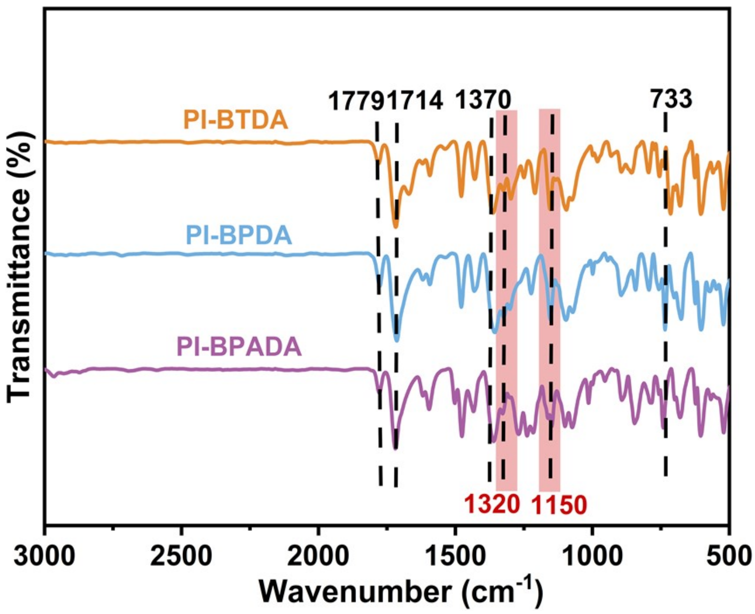


Fig. S1. FTIR spectra of the synthesized PI binders.

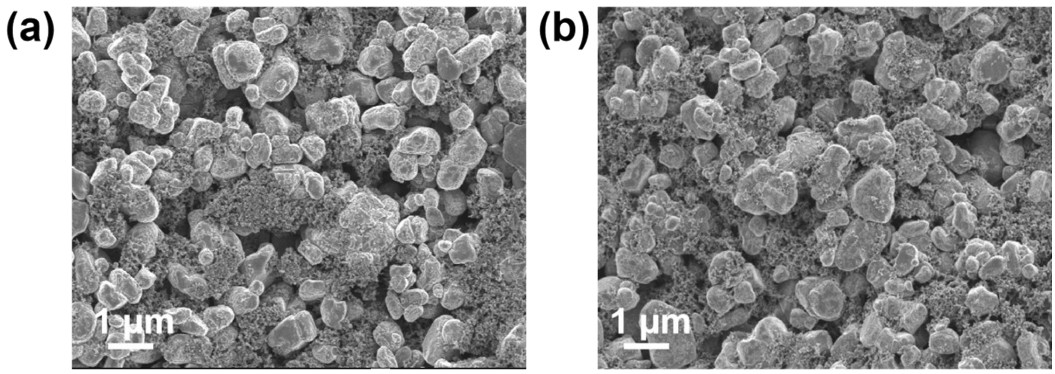


Fig. S2. SEM images of initial NCM811 Cathodes with (a) PI-BPDA binder and (b) PI-BTDA binder.

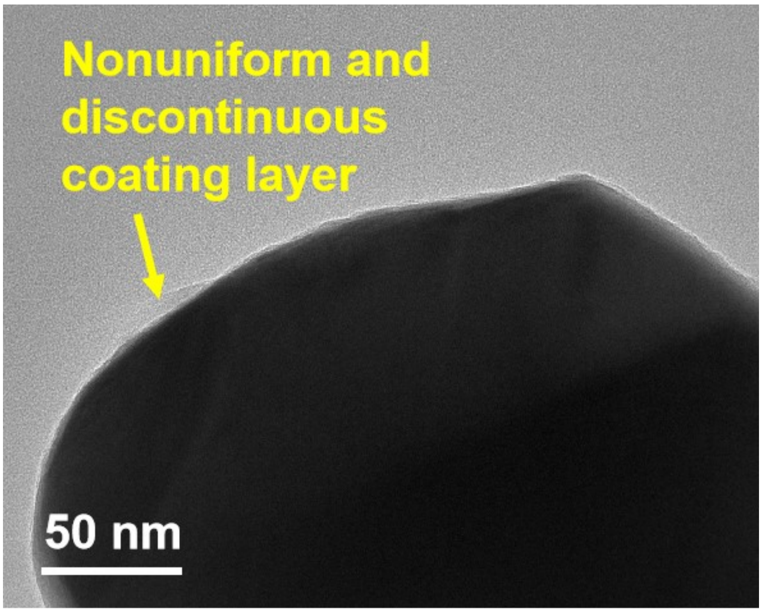


Fig. S3. The TEM image with low-magnification and large view files of initial PVDF/NCM cathode.

Table S1

Peel strength of NCM811 cathodes using PVDF or PI as binders

Materials	Average force (N)	Peel strength ($N\ mm^{-1}$)
PI-BPADA/NCM	9.28	0.42
PI-BPDA/NCM	8.55	0.39
PI-BTDA/NCM	10.86	0.42
PVDF/NCM	7.31	0.24

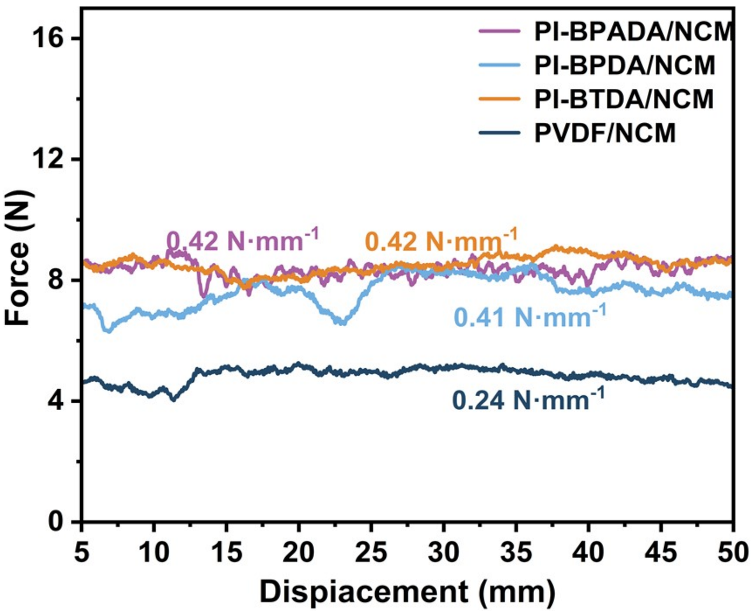


Fig. S4. Peel strength of NCM811 cathodes with PVDF/MCM and PI/NCM.

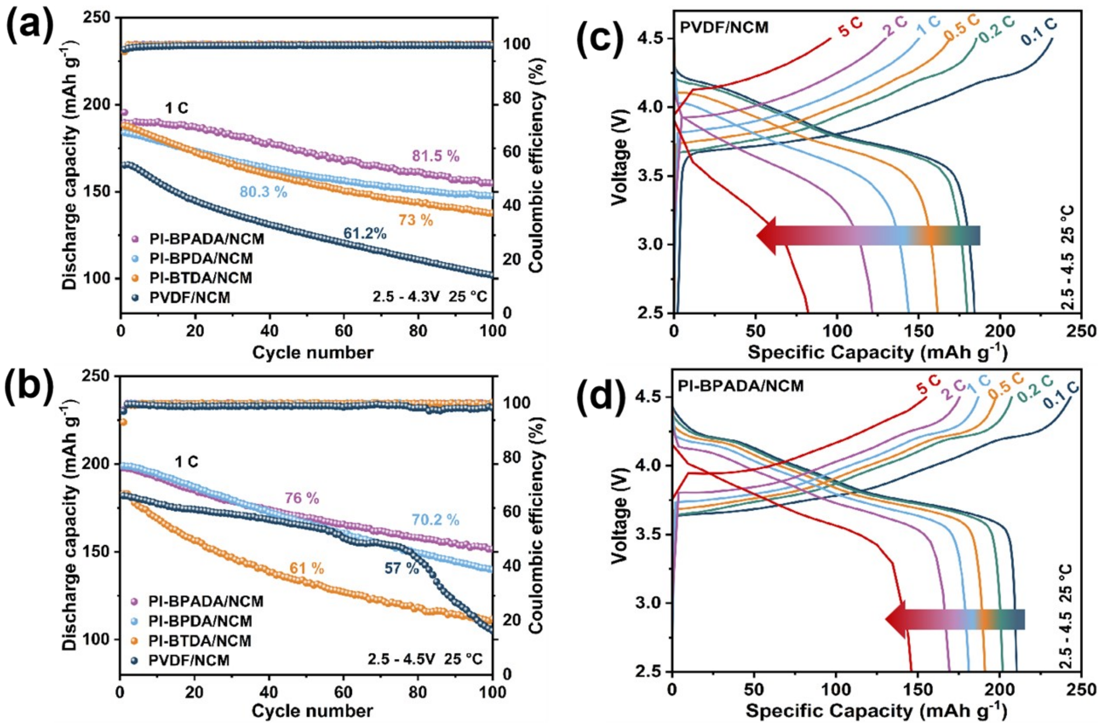


Fig. S5. Cycling performance of NCM811 cathodes with different binders after 100 cycling at 1C in the voltage range of (a) 2.5-4.3 V and (b)2.5-4.5 V. Voltage profiles corresponding rate performance for different cycles in the voltage range of 2.5-4.3 V, (c) PVDF/NCM and (d) PI-BPADA/NCM cathodes.

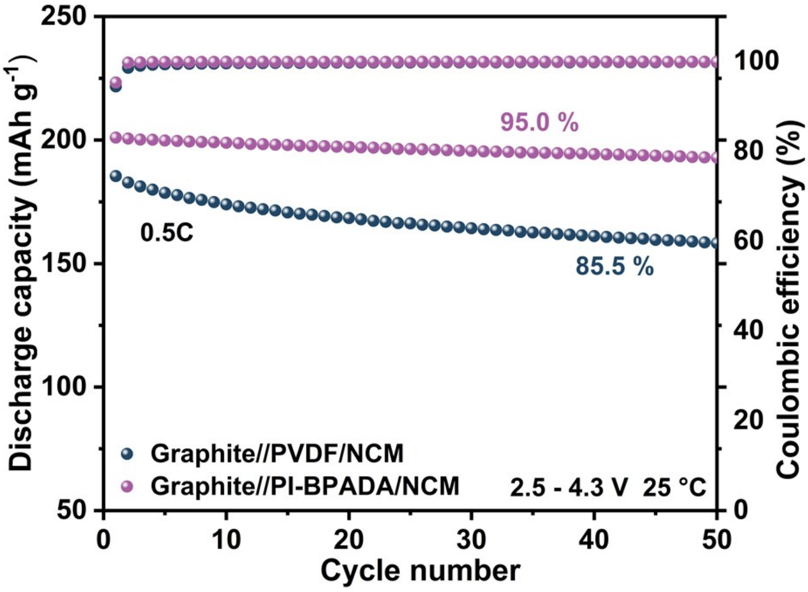


Fig. S6. Cycling performance of graphite||NCM811 full cells employing PVDF or PI-BPADA as the cathode binder

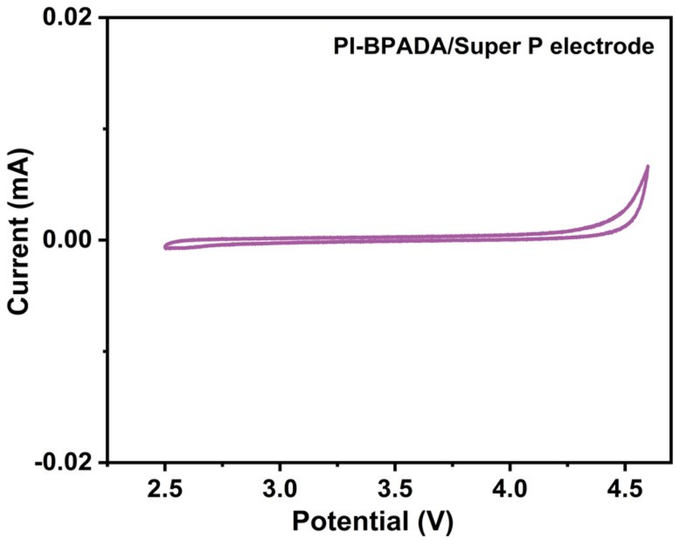


Fig. S7. CV Curve of the cathode with PI-BPADA + Super P on Al current collector with a scan rate of 0.1 mV s^{-1}.

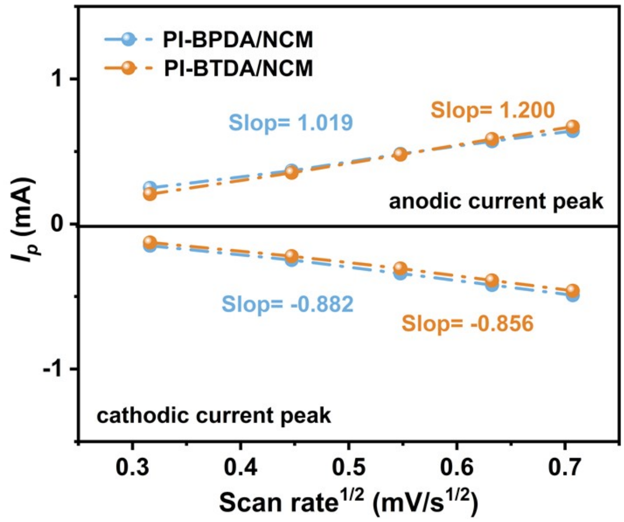


Fig. S8. Linear response of the peak current density (I_p) as a function of square root of scan rate for PI-BPDA/NCM cell and PI-BTDA/NCM cell.

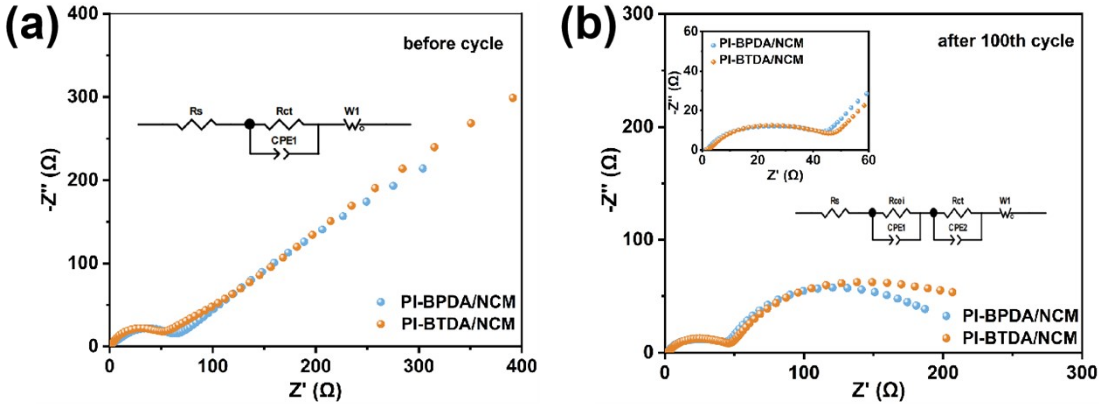


Fig. S9. Nyquist plots of PI-BPDA/NCM and PI-BTDA/NCM (a) at the initial state and (b) after 100 cycles in the voltage range of 2.5-4.3 V at 0.2C; insets are equivalent circuit models for fitting impedance spectra of initial and cycled cells.

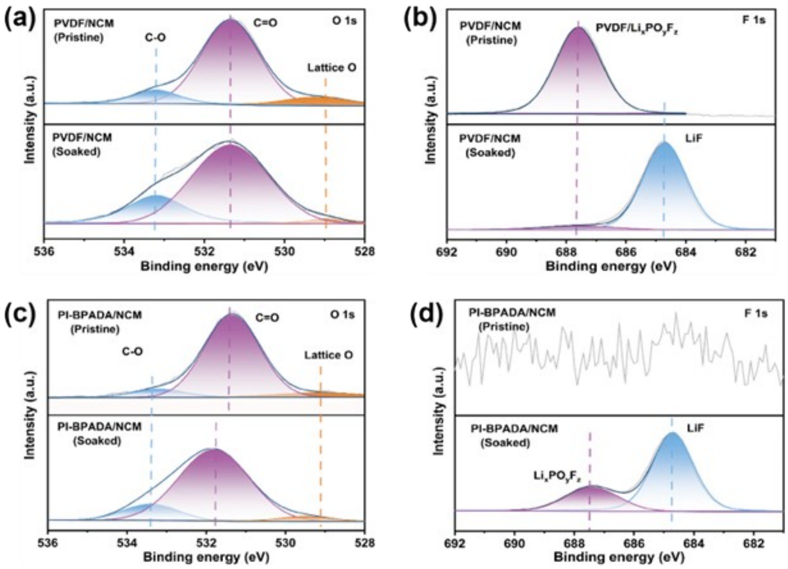


Fig. S10. O 1s and F 1s XPS spectra of (a,b) PVDF/NCM and (c,d) PI-BPADA/NCM electrodes for pristine and after soaking in electrolyte for 48h.

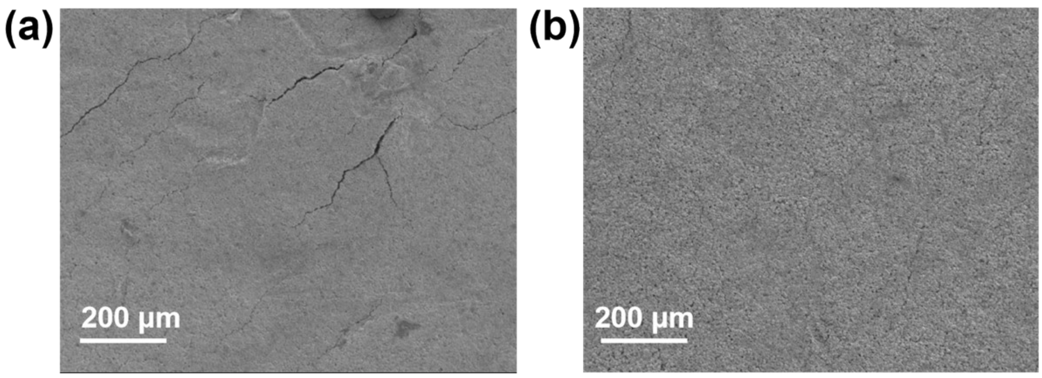


Fig. S11. SEM images of the (a) PVDF/NCM cathode and (b) PI-BPADA/NCM cathode after 100 cycles of cell in the voltage window of 2.5-4,3V.