Supplementary Materials for

The integration of $LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$ coatings on separators for elevated battery performance

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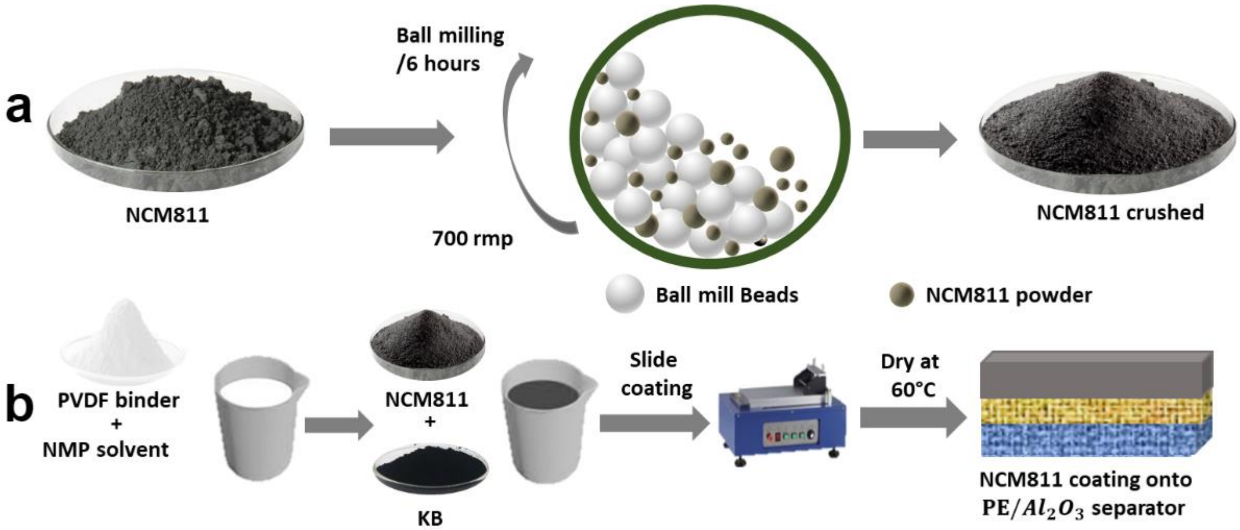

Figure S1 Schematic illustrating the crushing of NCM particles through planetary ball mill. (b) Illustration of the preparation of the triple layer separator

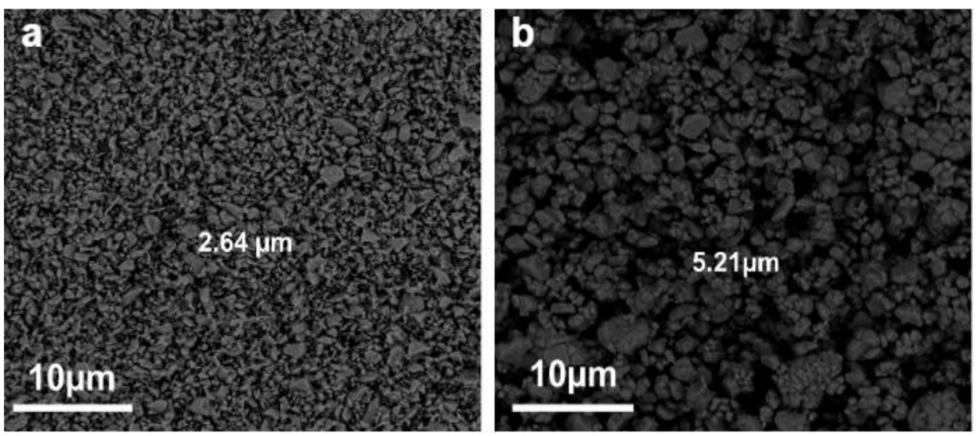

Figure S2 (a) SEM images the crushing of NCM particles through planetary ball mill. (b) the SEM image of the original unmilled NCM111 particles.

We have performed energy dispersive X-Ray spectroscopy (EDX) mapping analysis to confirm the presence and distribution of NCM811 coating on the PE/Al_2O_3 separator. The results in Figure S3 show the presence of key elements: The detection of Ni, Co, Mn and O signals confirms the successful coating of NCM811 particles on the separator surface. The Al and O signals indicate the presence of the Al_2O_3 ceramic layer. The C signals come from both the PE substrate and PVDF binder. The F signals originate from the PVDF binder used to adhere the coating layers. The EDX mapping demonstrates uniform distribution of all these elements, confirming that the NCM811 active material was successfully and homogeneously coated onto the PE/Al_2O_3 separator.

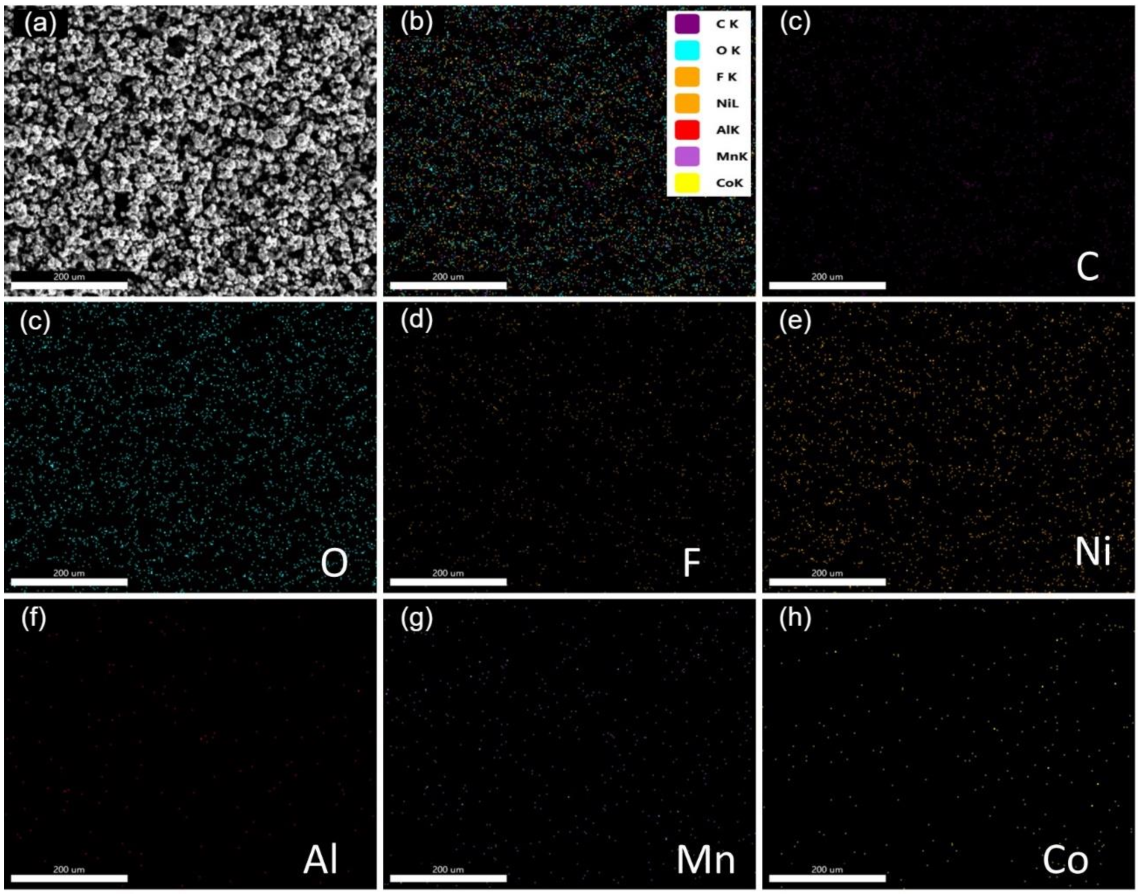

Figure S3 Structural characterizations for the NCM-crushed coated separator. (a) SEM image of NCM-crushed coated separator, (b-h) EDX mapping of NCM-crushed coated separator of different elements.

Figure S4(a) shows the original CV curves without normalization; Figure S4(b) presents the CV curves normalized by the mass loading on the cathode only, this accounts for slight variations in cathode mass between cells; Figure S4(c) shows the CV curves normalized by the total active material mass, including both the cathode and the NCM coating on the separator. This provides the most comprehensive normalization, as it accounts for all electrochemically active material in the cell. By normalizing to mA/g, we can now directly compare the specific currents between different separator configurations while eliminating the effect of mass loading variations. This normalization reveals that: the NCM-coated separators show higher specific peak currents compared to the uncoated separators, indicating improved utilization of the active material, the integrated area of the CV curves, which correlates to capacity, is larger for the NCM-coated separators on a per-gram basis. These normalized results provide stronger evidence for the benefits of the NCM coating on separator performance.

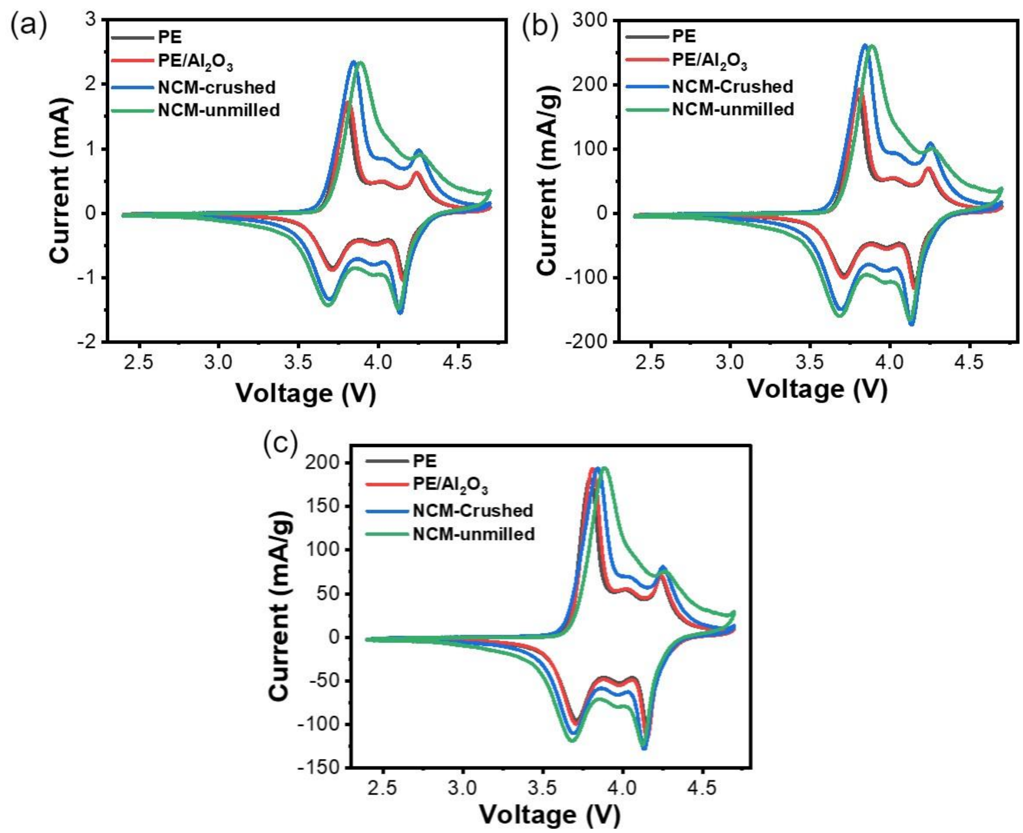

Figure S4 Cyclic voltammetry (CV) profiles of NCM811/Li half-cells with different separators (a) without any consideration of mass load, (b) normalized to mass load on the cathode, (c) normalized to mass load on the cathode and the separator.

To better demonstrate the electrochemical reaction of NCM811 coating on the separator, we have conducted the impedance analysis, charge-discharge tests and cycling performances for cells with three different configurations:

(1) Li||PE/Al_2O_3||NCM811: A half-cell with PE/Al_2O_3 (NCM811 coating on Al);

(2) Li||PE/Al_2O_3/NCM811||Al: A half-cell with PE/Al_2O_3/NCM811 separator but only Al current collector was used at the cathode side (NCM coating on PE/Al_2O_3);

(3) Li||PE/Al_2O_3/NCM811||NCM811: A half-cell using PE/Al_2O_3/NCM811 separator and NCM811 cathode (NCM811 coating on both separator and Al).

All PE/Al_2O_3/NCM811 separators used here are NCM-10 separator as mentioned in the manuscript. Figure S5(a) illustrate the impedance characteristics of different cells. The charge transfer resistance follows the order: Li||PE/Al_2O_3/NCM811||NCM811 < Li||PE/Al_2O_3/NCM811||Al < Li||PE/Al_2O_3||NCM811, indicating that the NCM811-coated separator can significantly decrease the transfer resistance.

Figure S5(b) shows that the first charge-discharge capacity profiles of different cells at 0.5 C. A clear charge/dis-charge plateau can be seen from 3.33 to 4.48 V for all cells, which is related to the electrochemical-extraction and insertion of Li-ions into NCM81. The result indicates that the NCM811 coating layer on PE/Al_2O_3 can participate in the electrochemical reaction and provide additional capacity.

The cyclic stability performances of different cells at 0.5 C are shown in Figure S5(c). The Li||PE/Al_2O_3/NCM811||Al cell and Li||PE/Al_2O_3||LFP cell have initial capacities of 205.0 mA h g^{-1} and 166.4 mA h/g, respectively. The coulombic efficiency Li||PE/Al_2O_3/NCM811||Al cell and Li||PE/Al_2O_3||LFP are 98.4 and 92.2%, respectively. The Li||PE/Al_2O_3/NCM811||NCM811 cell exhibits higher initial capacity of 216.39 mAh/g, indicating that the interface effect between NCM811 coating on separator and the NCM811 cathode plays a critical role in the significantly increased capacity.

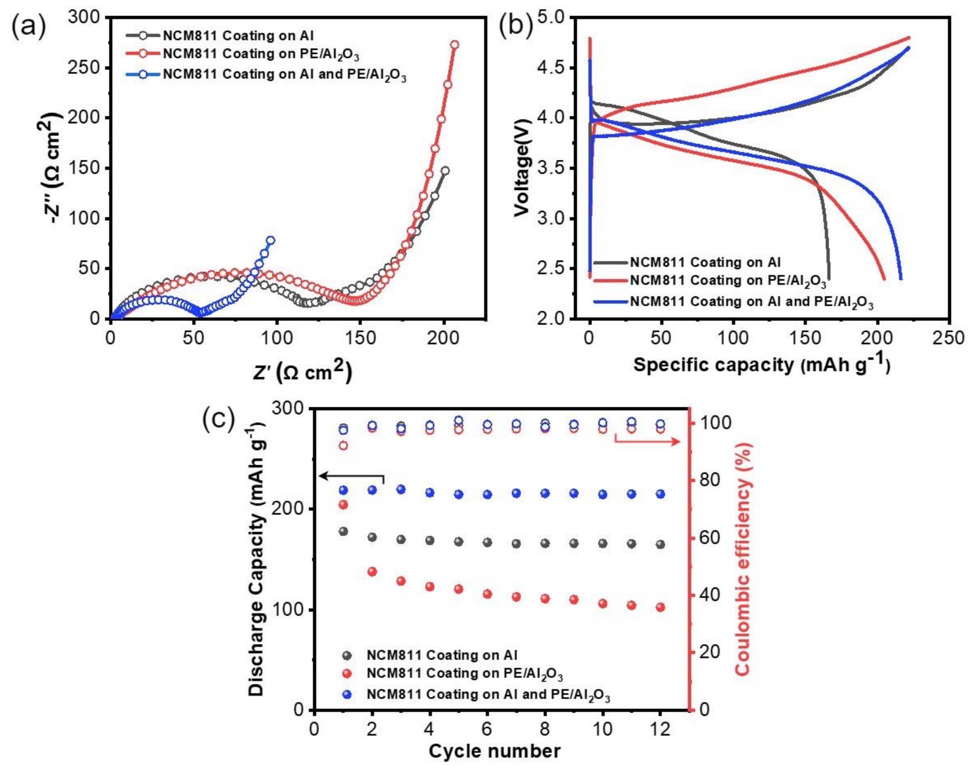

Figure S5 (a) Nyquist plot of the cells with the configuration Li||PE/Al_2O_3/NCM811||Al (NCM811 was coated on the PE/Al_2O_3 separator), Li||PE/Al_2O_3||NCM811 (NCM811 was coated on Al foil), and Li||PE/Al_2O_3 /NCM811||NCM811 (NCM811 was coated on both PE separator and Al foil) at 0.5 C. (b) Charge/discharge curves at 0.5 C. (c) Cyclic stability performances at 0.5 C.

PE separators are relatively inexpensive due to mature manufacturing processes; Al_2O_3 coatings add minimal cost due to the low price of alumina; NCM811 is significantly more expensive than PE or Al_2O_3, increasing material costs. As shown in Table S1, coating NCM811 or other active ceramic layer on PE/Al_2O_3 separator incurs active ceramic material cost, and additional labor/energy consumption/depreciation cost. If the active coating on the separator fully participates in the charge/discharge reaction (100% utilization), the active material cost on the separator is equivalent to the cost caused by the increase in cathode active material. In this case, the cost for NCM coated separator can be solely attributed to labor/energy consumption/depreciation cost, about 0.56% cost increase compared to PE/Al_2O_3. If the utilization rate of active ceramics is 90%, the cost increase ratio per square meter will be 1.56%.

Table S1 Estimated cost for active ceramic coated separators

Separator coating ceramic	**LFP**	**NCM**	**Graphite**	**LTO**
1 GWh separator usage (m^2)	15000000	15000000	15000000	15000000
Yield rate	70%	70%	70%	70%
Coating thickness (um)	5	5	5	5
Density of ceramic (g/cm^3)	1.5	2.9	1.7	1.8
Coating mass per square (g/m^2)	7.5	14.5	8.5	9.0
Coating mass for 1GWh cell (ton)	112.5	217.5	127.5	135
Ceramic price (RMB Yuan/ton)	17	35	5	11
Labor/energy consumption/depreciation cost (RMB Yuan/ m^2)	0.30	0.30	0.30	0.30
Ceramic cost (RMB Yuan/ m^2)	1.28	5,98	0.43	0.99
Total coating cost (RMB Yuan/m^2) (90% utilization rate of active ceramics)	0.43	0.90	0.34	0.40
Coating cost (RMB million Yuan/GWh) (90% utilization rate of active ceramics)	6.45	13.50	5.10	6.00
Cost increase ratio per square (%) (90% utilization rate of active ceramics)	0.81%	1.69%	0.64%	0.75%
Cost per square (RMB million Yuan/m^2) (100% utilization rate of active ceramics)	0.30	0.30	0.30	0.30
Cost increase ratio per square (%) (100% utilization rate of active ceramics)	0.56%	0.56%	0.56%	0.56%

Note: LFP: $LiFePO_4$, NCM: $LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$, LTO: $Li_4Ti_5O_{12}$, some price estimate data are from Guangfa Securities of China.