Supporting Information

Plasma-Assisted Mechanochemistry to Covalently Bond Ion-Conducting Polymers to Ni-Rich Cathode Materials for Improved Cyclic Stability and Rate Capability

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SEM images pristine NCM811 particles and pristine PVDF particles. SEM images with EDS mapping of the PMC-processed composite without plasma treatment before and after the filtration. STEM images of PMC-filtered composites. TGA curve of pristine NCM811 and PMC-processed composites without plasma treatment after filtration. XPS spectra of the PMC+filtered, mixed NCM/PVDF composites and pristine PVDF, NCM811 particles. Initial charge/discharge curves of 3% and 20% mixed and PMC-processed and PMC-filtered NCM/PVDF electrode at 0.5 C. EIS spectra and electrochemical properties (including Warburg coefficient) of simply mixed and PMC-processed electrodes before and after 100 cycles. EDS elemental mapping images of lithium metal anode with PMC-filtered and simply mixed NCM cathodes.

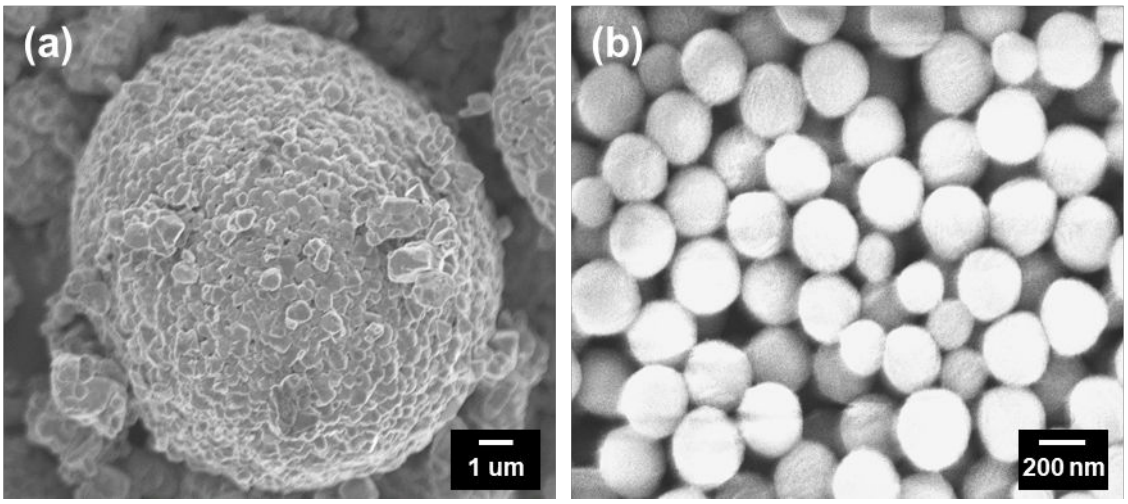


Figure S1. SEM images of (a) pristine NCM811 particles and (b) pristine PVDF particles.

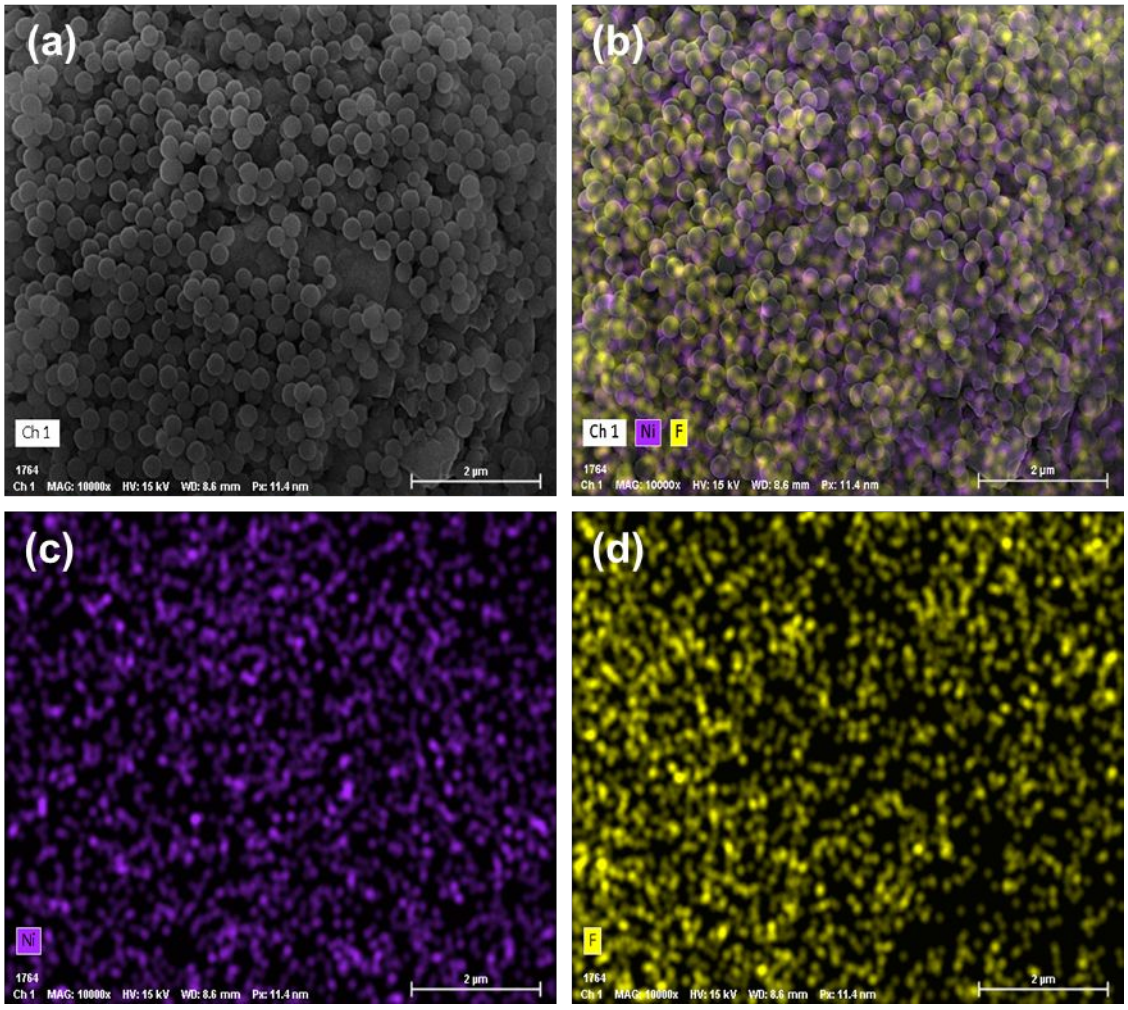


Figure S2. SEM images (a) with EDS mapping (b-d) of the PMC-processed composite without plasma treatment.

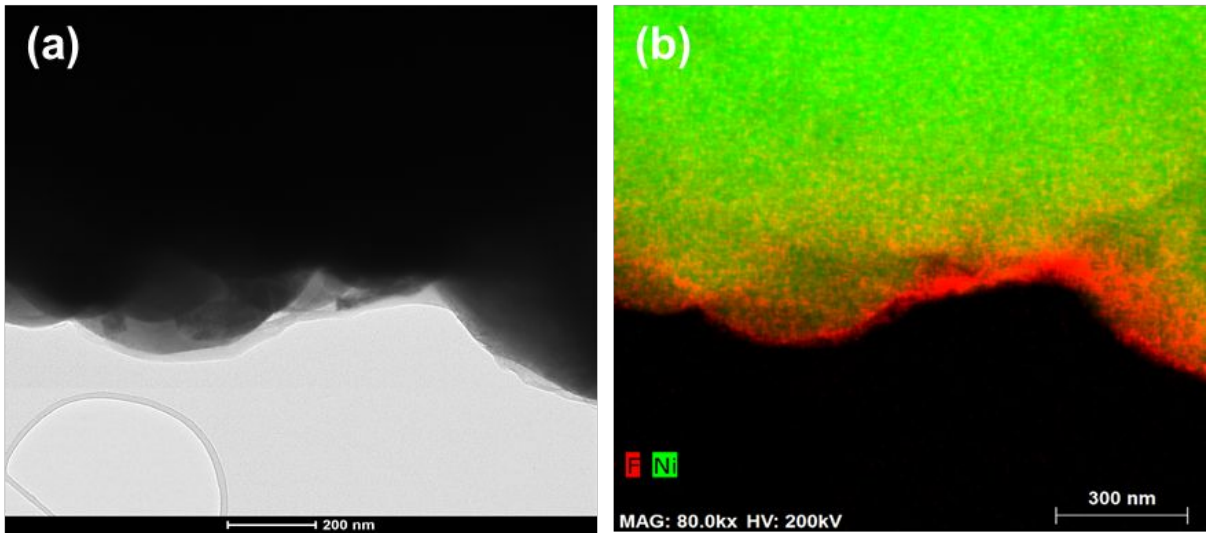


Figure S3. STEM images of PMC-filtered composites.

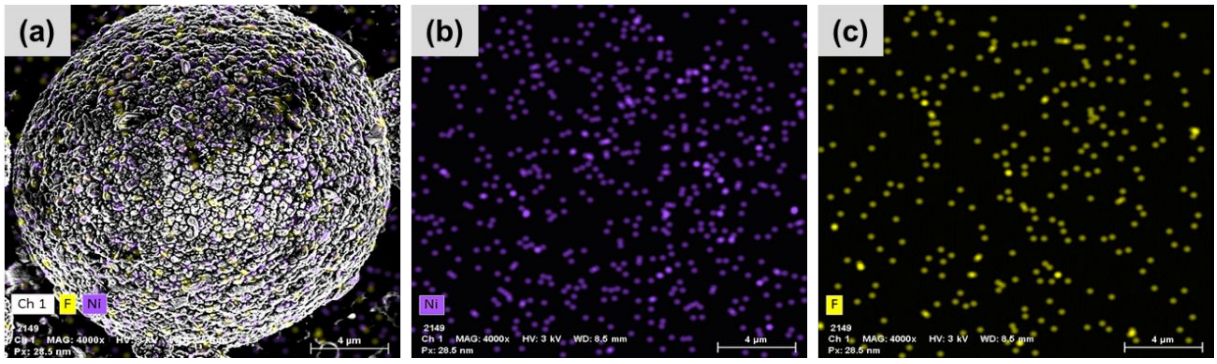


Figure S4. SEM images (a) with EDS mapping (b-e) of the PMC-processed composite without plasma treatment after filtration.

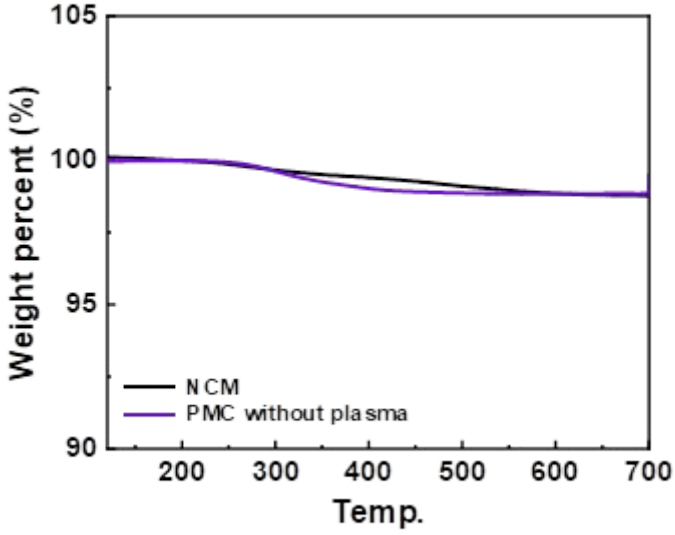


Figure S5. TGA curve of pristine NCM811 and PMC-processed composites without plasma treatment after filtration.

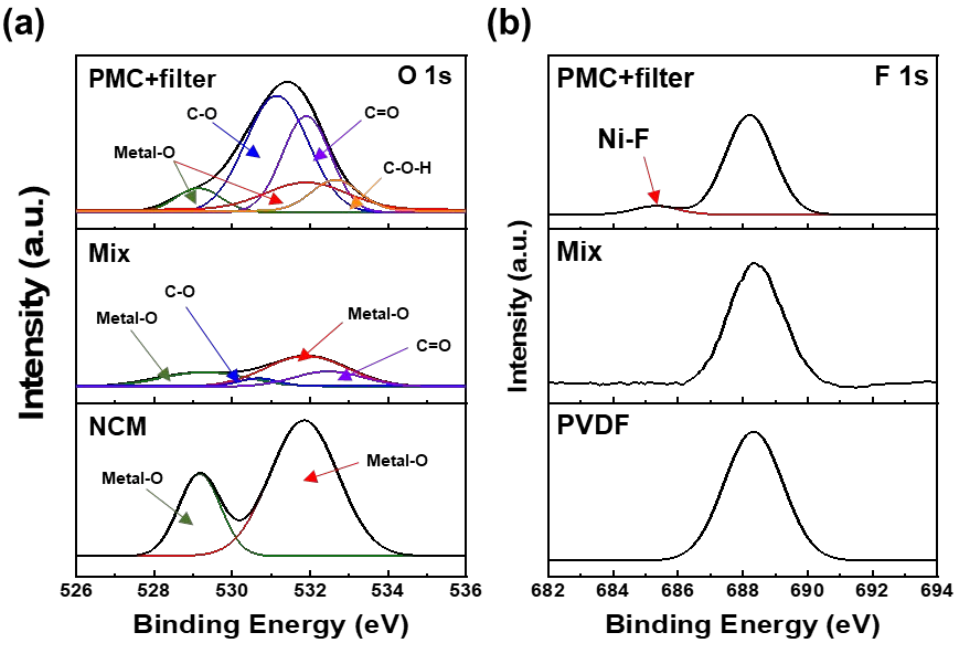


Figure S6. XPS spectra of the samples (PMC+filtered, mixed NCM/PVDF composites and pristine PVDF, NCM811 particles): (a) O1s and (b) F1s.

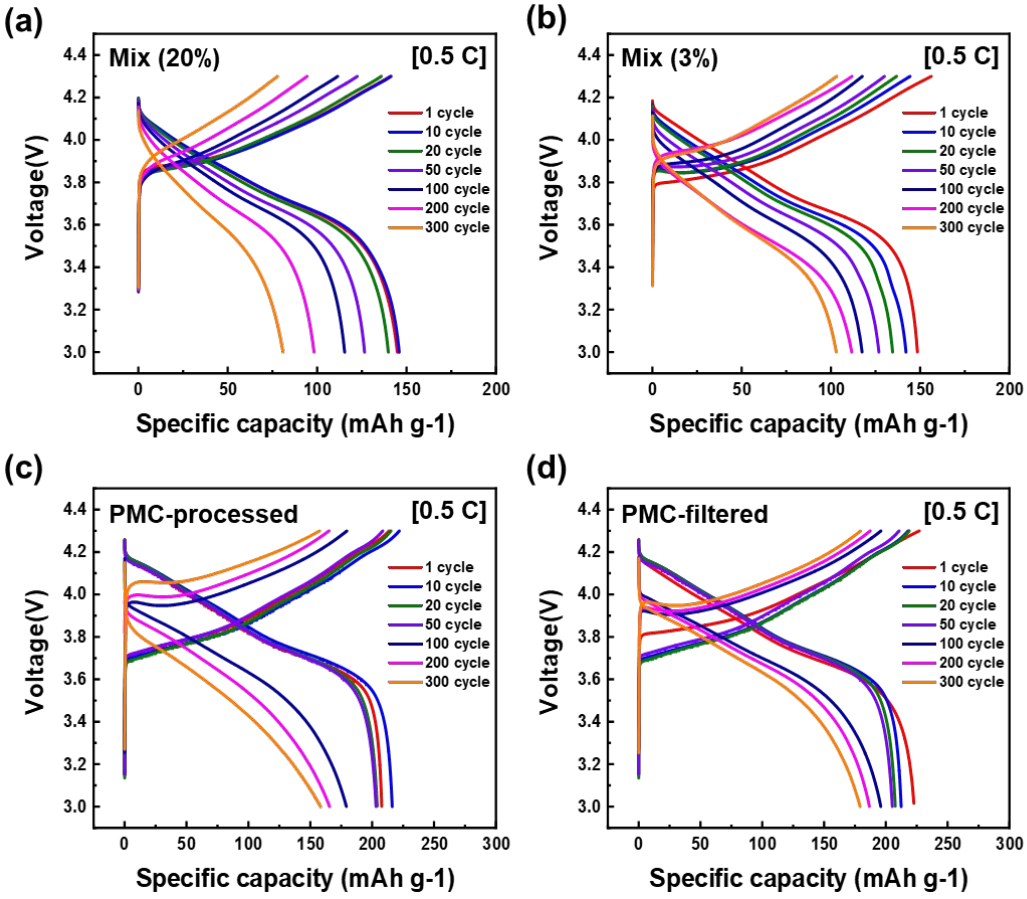


Figure S7. Initial charge/discharge curves of (a) mixed (3% of actual binder content) and (b) mixed NCM/PVDF electrode (20% of actual binder content) at 0.5 C. Initial charge/discharge curves of (c) PMC-processed and (d) PMC-filtered NCM/PVDF electrode at 0.5 C.

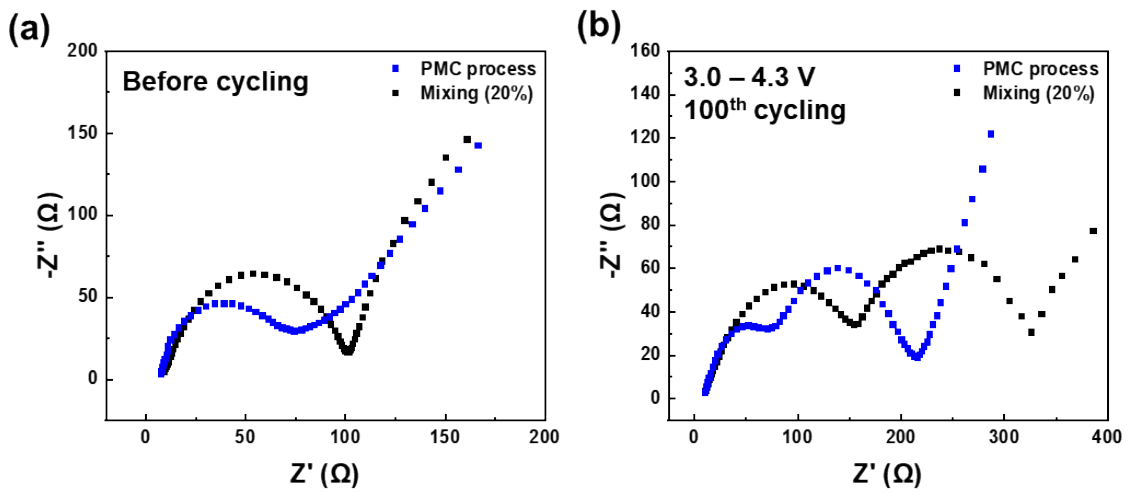


Figure S8. (a and b) EIS spectra of simply mixed (20 wt%) and PMC-processed electrodes measured at 25 °C (a) before and (b) after 100 cycles of charging/discharging.

Table S1. The electrochemical properties of simply mixed (20 wt%) and PMC-processed NCM/PVDF electrodes were obtained from impedance spectroscopy before and after 100 cycles.

Sample	Cycles	R_b (Ω)	R_{SEI}(Ω)	R_{ct} (Ω)
Mix (20%)	**0th**	9.2	-	91.1
	100th	10.7	143.6	172.7
PMC	**0th**	8.0	-	64.2
	100th	10.3	61.4	141.8

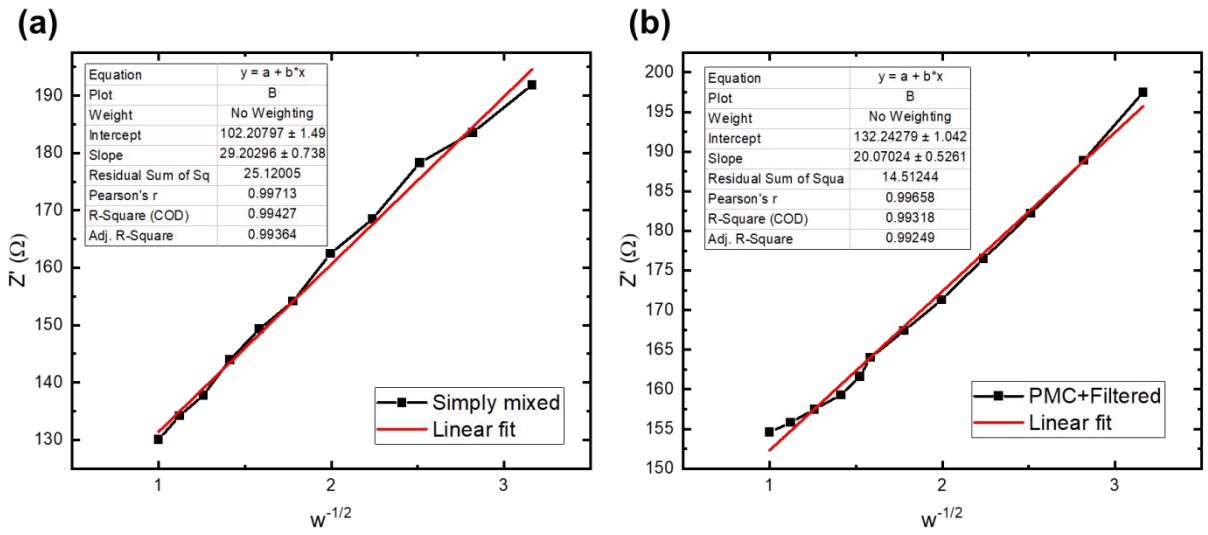
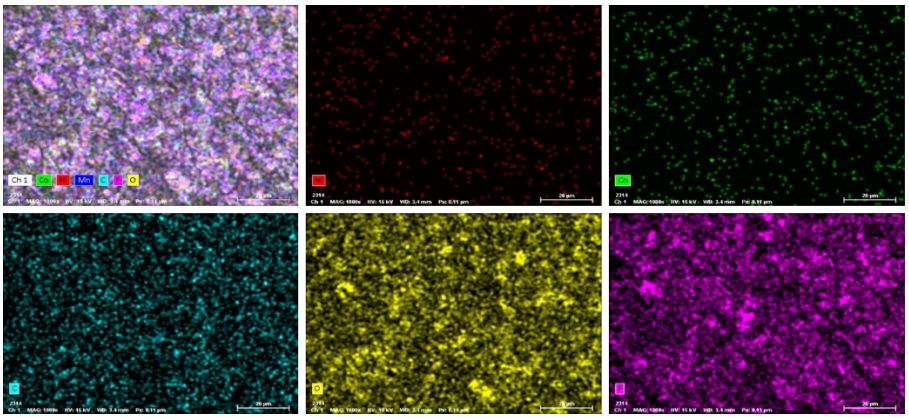


Figure S9. The linear relationship between the Warburg impedance and the inverse square root of angular frequency, the slopes of the simulated lines are the Warburg coefficient (σ) for (a) simply mixed (3 wt%) electrode and (b) PMC filtered electrode after 100 cycles. The lithium-ion diffusion coefficient (D_{Li^+}) was calculated using the data from the impedance spectrum according to the following equation.

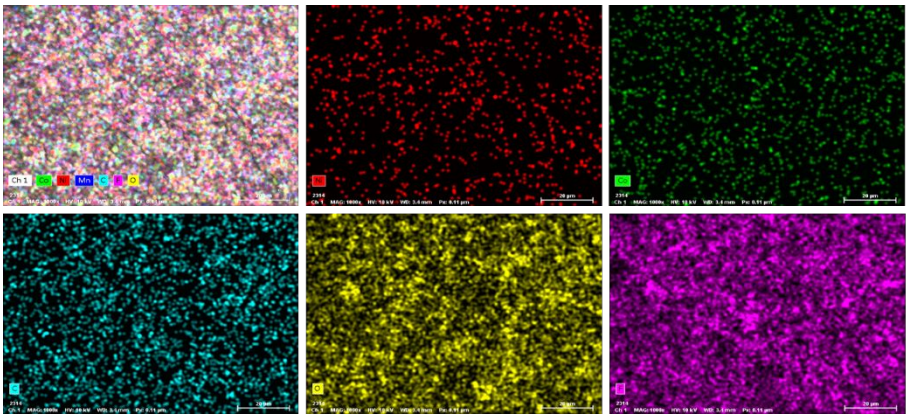
$$D_{Li^+} = \frac{R^2T^2}{2A^2n^4F^4c^2\sigma^2}$$

In this equation, R is the gas constant, T is the Kelvin temperature, A is the geometric area of the cathode, n is the number of electrons transferred in the redox process, F is the Faraday constant, and c is the concentration of lithium ions. The σ is the Warburg coefficient, which can be obtained from the slope of the straight line between Z' and $\omega^{-1/2}$ axes.



Element	At. No.	Mass [%]	Mass Norm. [%]	Atom [%]
Ni	28	1.94	1.94	0.59
Co	27	2.44	2.44	0.74
Mn	25	0.27	0.27	0.09
C	6	9.55	9.55	14.11
O	8	24.76	24.76	27.46
F	9	61.05	61.05	57.02
		100.00	100.00	100.00

Figure S10. EDS elemental mapping images of lithium metal anode with PMC-filtered NCM cathode after 100 charging-discharging cycles at 0.5 C.



Element	At. No.	Mass [%]	Mass Norm. [%]	Atom [%]
Ni	28	3.14	3.14	0.96
Co	27	4.32	4.32	1.32
Mn	25	0.00	0.00	0.00
C	6	11.36	11.36	17.00
O	8	22.19	22.19	24.93
F	9	58.99	58.99	55.80
		100.00	**100.00**	**100.00**

Figure S11. EDS elemental mapping images of lithium metal anode with simply mixed NCM cathode after 100 charging-discharging cycles at 0.5 C.