

Supporting Information

Electrophoretically deposited artificial cathode electrolyte interphase for improved performance of NMC622 at high voltage operation

Inbar Anconina and Diana Golodnitsky

School of Chemistry, Tel Aviv University, Tel Aviv 6997801, Israel

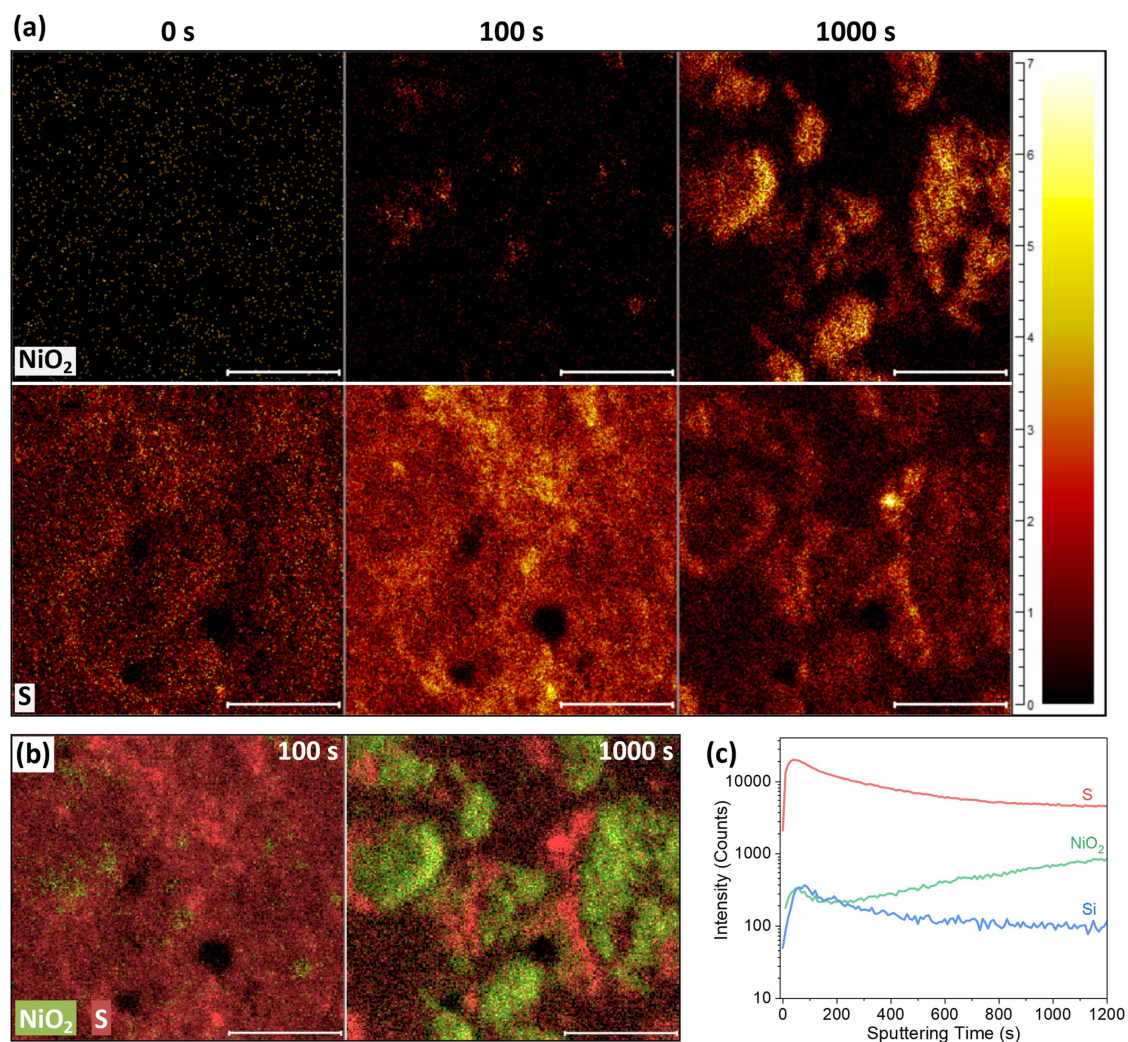


Fig. S1 Negative TOF-SIMS mapping images of the LPSC-coated NMC622 (scale bar: 10 μm), display the individual mapping signals for NiO_2 and S at different sputtering times: 0, 100, and 100 seconds (a); and their merged images for sputtering times of 100 and 1000 seconds (b). (c) The depth profiles of NiO_2 , S, and Si, indicating the presence of the NMC cathode, LPSC, and PIL polymer, respectively.

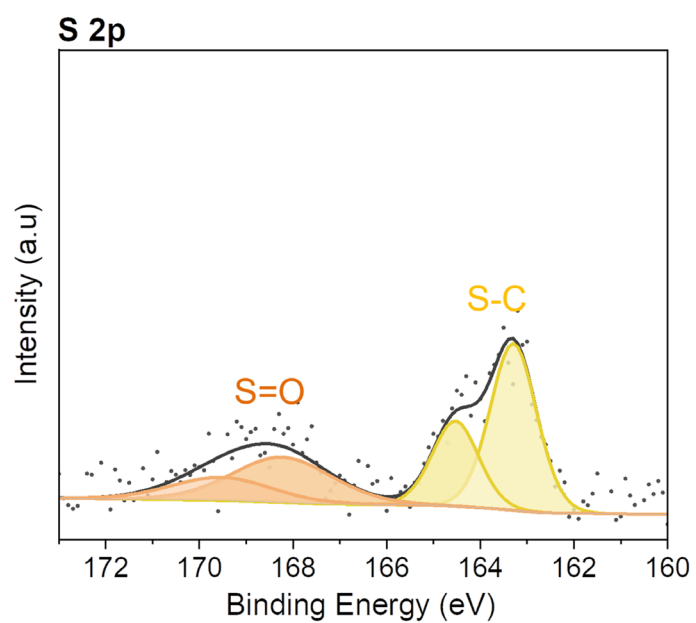


Fig. S2 S 2p XPS spectrum of pristine PIL (PDMS-g-MPA·Li⁺₁₃/PEGMEMA₂₇).

Table S1 Summary of the electrochemical performance during first and second charge-discharge cycle of NMC622 cathodes versus Li electrodes at 30 °C and cycling rate of C/15.

NMC622/Li	Charge capacity [mA/g]	Discharge capacity [mA/g]	Coulombic Efficiency [%]	ICL [%]
4.3V cutoff voltage				
First cycle				
Pristine	206.8	190.8	92.3	7.7
LPSC-coated	218.8	199.2	91.0	9.0
Second cycle				
Pristine	191.2	190.9	99.8	0.16
LPSC-coated	200.4	199.8	99.7	0.30
4.5V cutoff voltage				
First cycle				
Pristine	233.8	215.2	92.0	7.9
LPSC-coated	248.0	228.0	91.9	8.1
Second cycle				
Pristine	216.7	215.4	99.4	0.60
LPSC-coated	230.6	229.2	99.4	0.61

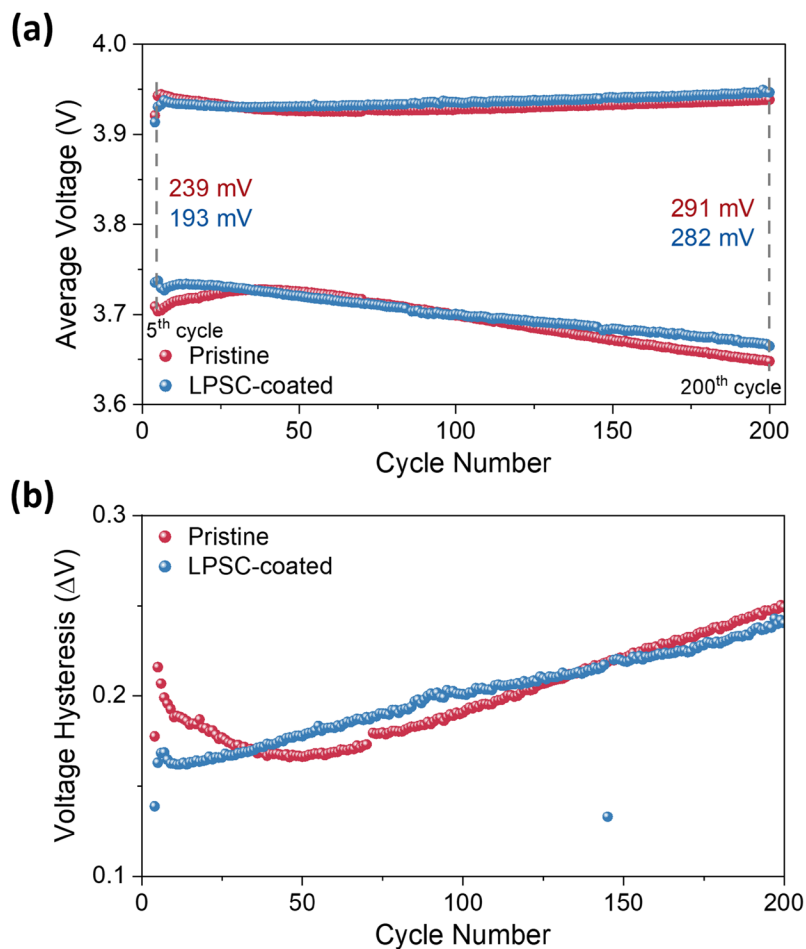


Fig. S3 (a) Average voltage profiles in charge and discharge and (b) voltage hysteresis calculated as the difference between average charge and discharge voltages for the pristine and LPSC-coated NMC622 cathodes at 4.3V cutoff voltage.

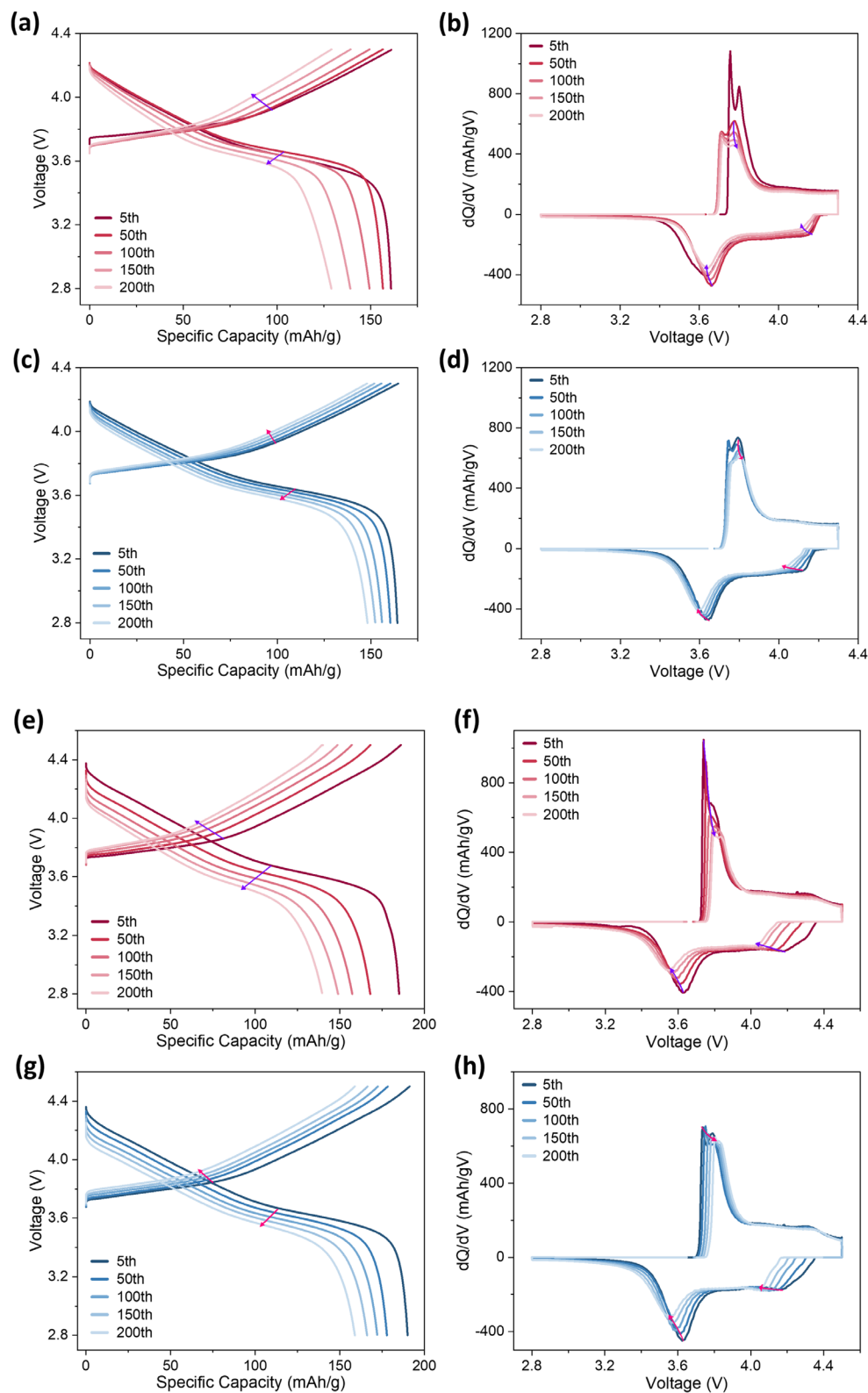


Fig. S4 Voltage profiles and corresponding differential capacity dQ/dV versus V plots for NMC622 cathodes comprising: (a,b)&(e,f) pristine, and (c,d)&(g,h) LPSC-coated, at the 5th, 50th, 100th, 150th, and 200th cycle, at 2.8-4.3 V (a-d) and 2.8-4.5 V (e-h) (0.5C-1.0 charge-discharge rates).

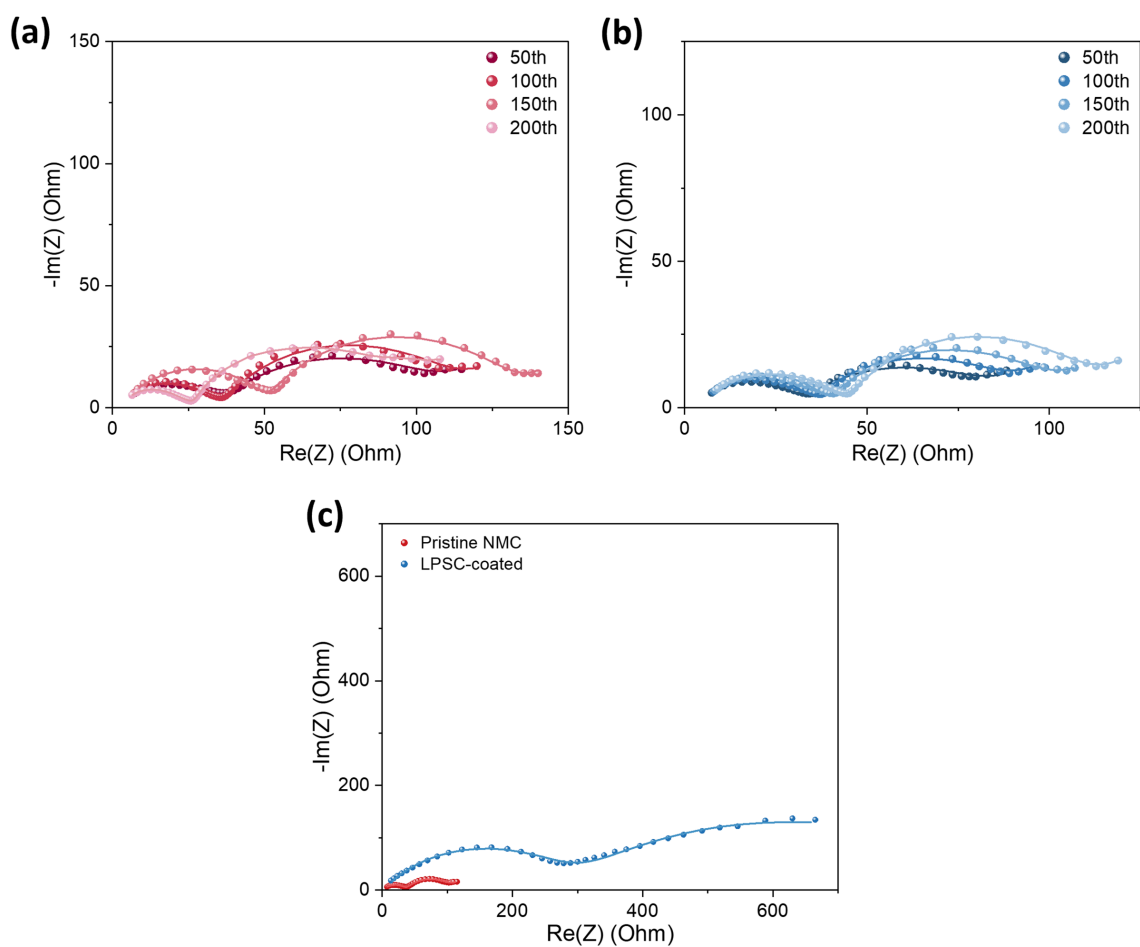


Fig. S5 Nyquist plots of EIS measurements and their fit during electrochemical cycling after the 50th, 100th, 150th, and 200th cycle at 2.8V-4.5V of pristine (a) and LPSC-coated (b) cells; and comparison of the measurements of the third cycle (c).

Table S2 Fitted impedance values by the corresponding equivalent circuits of pristine and LPSC-coated NMC622 cathodes and the corresponding estimated lithium-ion diffusion coefficients (D_{Li}).

Cycle number	$R_1=R_{bulk}$ [Ohm]	$R_2=R_{SF}$ [Ohm]	$C_2=C_{SF}$ [F]	$R_3=R_{CT}$ [Ohm]	$C_3=C_{CT}$ [F]	W [Ohm.s ^{0.5}]	χ^2 [Ohm ²]	$\chi^2/[Z^2]$	D_{Li} [cm ² .s ⁻¹]
Pristine NMC622									
3rd	1.1	36.2	3.12E-6	66.5	5.40E-3	2.68	15.3	8.12E-3	2.464E-14
50th	1.3	34.1	2.85E-6	71.8	5.40E-3	2.52	1.42	5.20E-3	2.767E-14
100th	1.5	37.7	1.89E-6	75.2	7.60E-3	2.57	3.03	3.92E-3	2.673E-14
150th	3.6	47.1	3.94E-6	79.5	4.38E-3	2.82	1.55	3.40E-3	2.212E-14
200th	1.1	24.0	2.30E-6	66.1	2.30E-3	4.22	5.32	4.24E-3	9.888E-15
LPSC-coated NMC622									
3rd	1.5	284.5	3.94E-6	551.8	11.98E-3	5.75	349	8.34E-3	5.354E-15
50th	2.1	28.6	2.93E-6	52.9	5.55E-3	1.98	3.79	6.14E-3	4.478E-14
100th	5.1	29.3	4.21E-6	53.0	6.03E-3	2.19	5.90	7.60E-3	3.675E-14
150th	5.1	33.8	3.87E-6	60.4	6.88E-3	2.36	6.71	8.06E-3	3.178E-14
200th	2.5	42.4	2.85E-6	63.0	7.53E-3	3.04	8.16	6.64E-3	1.911E-14

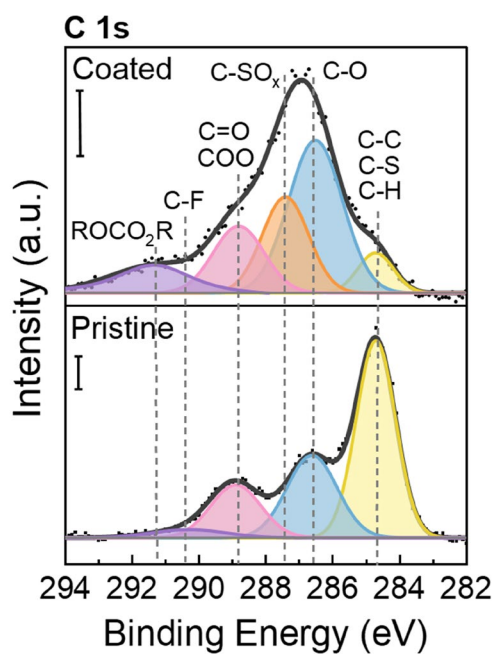


Fig. S6 C 1s XPS spectra in the CEI of pristine and LPSC-coated NMC622 after 20 cycles (200 s sputtering).

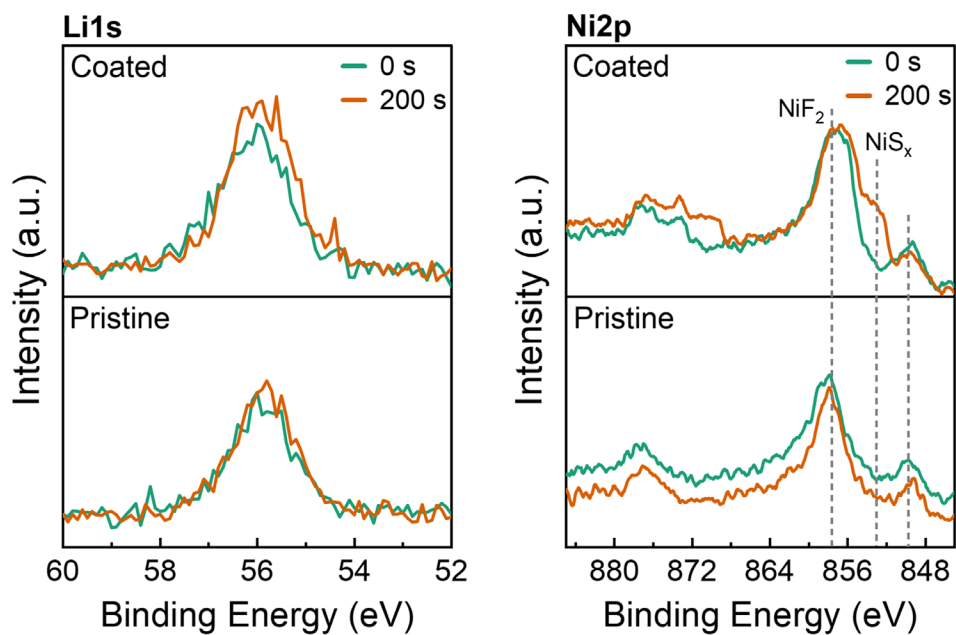


Fig. S7 Li 1s and Ni 2p XPS spectra of the SEI of Li metal anodes in pristine and LPSC-coated NMC622 cells after 20 cycles (surface and depth profiles after a 200 s sputtering).

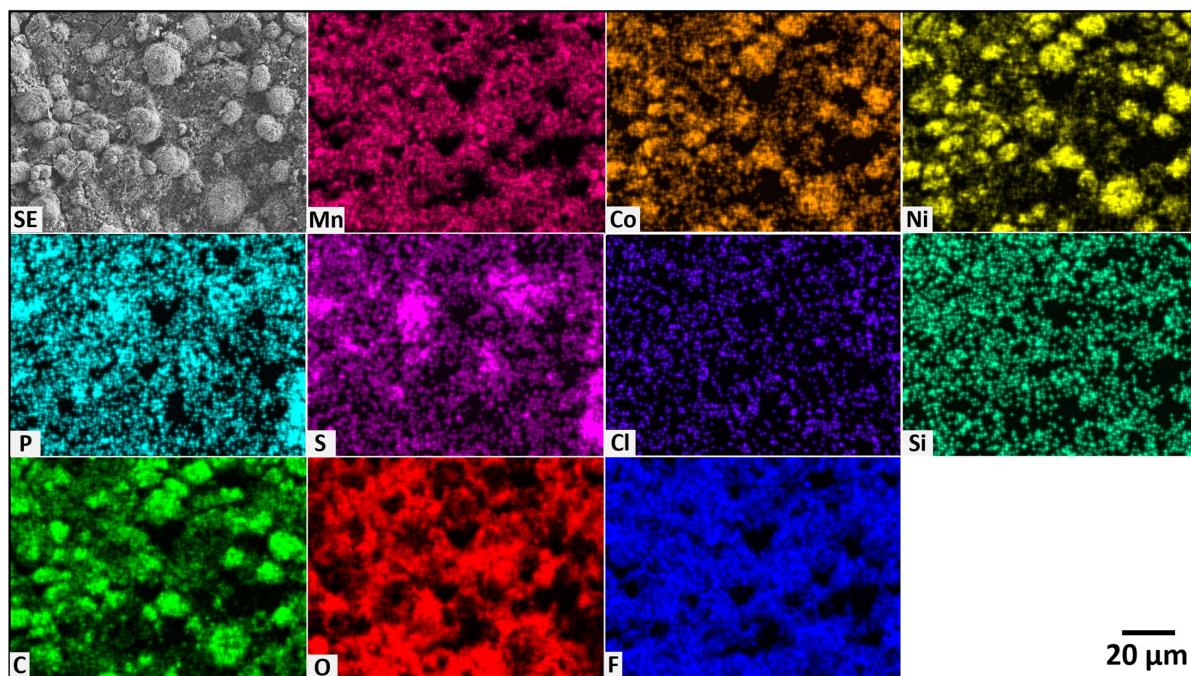


Fig. S8 SEM and EDS mapping images of cycled LPSC-coated NMC622 (after 200 cycles).