Probing The Electrode/Electrolyte Interface in The Lithium Excess Layered Oxide Li_{1.2}Ni_{0.2}Mn_{0.6}O₂

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Figure S1. Electron Energy Loss spectroscopy of samples at various points in the second cycle.



Figure S2. (a) F1s and (b) P2p region scans during the first electrochemical cycle.



Figure S3. X-ray photoelectron spectroscopy spectra of second cycled samples at various points.



Figure S4. Nickel K edge transmission XANES spectra during the first electrochemical cycle.



Figure S5. Chi-K data for pristine, 4.8V, 2.0V and 10 cycled electrodes for both transmission and electron yield mode XAS.

Cycle #	Sample Position	Cutoff Voltage	Capacity	
	В	Charge: 4.40 V	101 mAh/g	
	С	Charge: 4.45 V	111 mAh/g	
	D	Charge: 4.60 V	276 mAh/g	
Cycle 1	Е	Charge: 4.80 V	304 mAh/g	
	F	Discharge: 4.00 V	44 mAh/g	
	G	Discharge: 3.30 V	173 mAh/g	
	Н	Discharge: 2.00 V	256 mAh/g	
	I	Charge: 4.40 V	200 mAh/g	
	J	Charge: 4.45 V	210 mAh/g	
	К	Charge: 4.60 V	230 mAh/g	
Cycle 2	L	Charge: 4.80 V	247 mAh/g	
	М	Discharge: 4.00 V	44 mAh/g	
	N	Discharge: 3.30 V	157 mAh/g	
	0	Discharge: 2.00 V	238 mAh/g	

Table S1. Capacity and sample positions for the first and second electrochemical cycle.

	Position/V	Lattice Oxygen	%	OH/Carbonate (eV)	%	Superoxide (eV)	%
		(eV)					
1 st Cycle	a) Pristine	529.8	20.50	531.8	79.50	-	-
	b) 4.4V	529.3	41.14	531.9	55.69	535.0	3.17
	c) 4.6V	529.8	31.26	531.7	59.17	535.4	9.57
	d) 4.8V	529.6	35.78	531.8	49.87	535.0	14.35
	e) 2.0V	529.4	31.77	531.9	55.69	535.5	12.54
2 ND Cycle	f) 4.4V	529.8	27.33	532.2	62.04	535.3	10.63
	g) 4.6V	529.7	30.85	532.0	56.22	535.0	12.94
	h) 4.8V	529.8	23.53	532.6	71.04	535.8	5.43
	I) 2.0V	530.1	26.19	532.3	59.05	545.4	14.76-

Table S2. Analyzed data from x-ray photoelectron spectroscopy.

Sample	Mn-O (Å)	Mn-M (Å)	R-factor
Pristine-T	1.87(8)	2.90(4)	0.003
Pristine-EY	1.88(6)	2.89(4)	0.002
4.8V-T	1.87(2)	2.88(6)	0.0005
4.8V-EY	1.90(5)	2.90(4)	0.002
2.0V-T	1.89(4)	2.92(6)	0.004
2.0V-EY	1.91(4)	2.91(8)	0.002
10 Cycle-T	1.89(6)	2.91(4)	0.001
10 Cycle-EY	1.92(4)	2.89(2)	0.002

Table S3. EXAFS calculated bond lengths for T-XAS and TY-XAS samples.