Supporting information

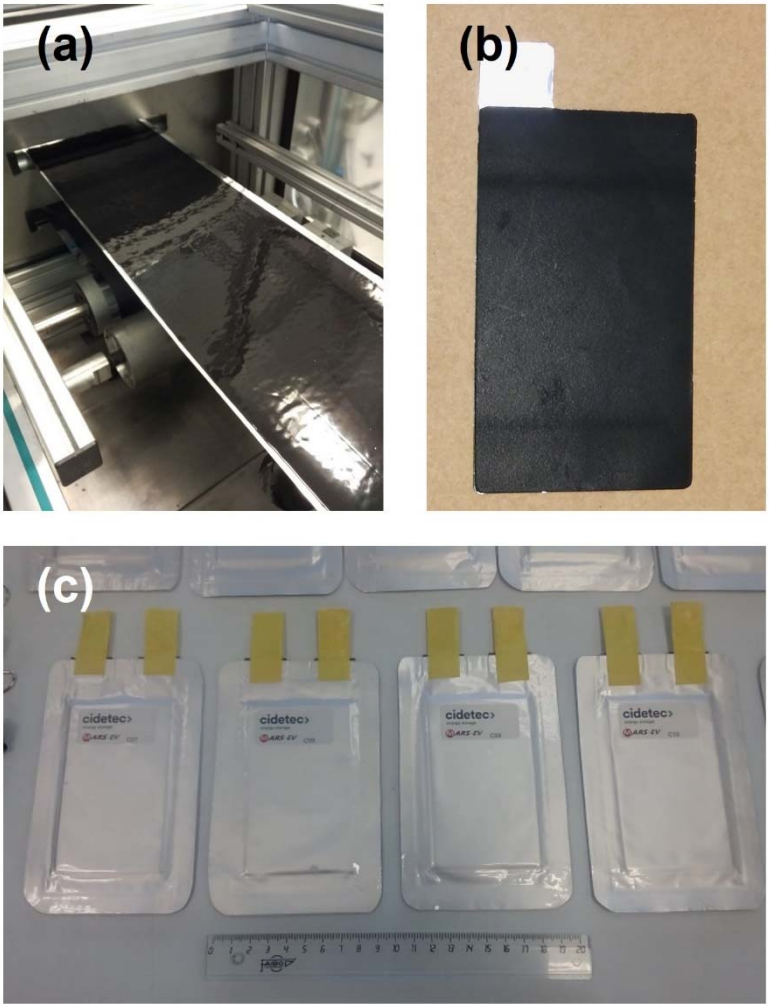

Figure S1. *Pictures of (a) the cathode coating after passing through the knife, (b) a cathode, and (c) assembled pouch cells.*

***Table S1**. Testing protocol applied to the LNMO HCCs.*

Step	Conditions	Cycles
Charge Discharge	0.2C → 5 V/CV < 0.05C & 2 s 0.2C → 3.5 V	3
Charge Discharge	0.5C → 5 V /CV < 0.05C & 2 s 0.5C → 3.5 V	1
Charge Discharge	1C → 5 V/CV < 0.05C & 2 s 1C → 3.5 V	3
Charge Discharge	0.5C → 5 V/CV < 0.05C & 2 s 0.5C → 3.5 V	1
Charge Discharge	1C → 5 V/CV < 0.05C & 2 s 2C → 3.5 V	3
Charge Discharge	0.5C → 5 V/CV < 0.05C & 2 s 0.5C → 3.5V	1
Charge Discharge	1C → 5 V/CV < 0.05C & 2 s 5C → 3.5 V	3
Charge Discharge	0.5C → 5 V/CV < 0.05C & 2 s 0.5C → 3.5 V	1
Charge Discharge	1C → 5 V/CV < 0.05C & 2 s 8C → 3.5 V	3
Charge Discharge	0.5C → 5 V/CV < 0.05C & 2 s 0.5C → 3.5 V	1
Charge Discharge	1C → 5 V/CV < 0.05C & 2 s 10C → 3.5 V	3
Charge Discharge	0.5C → 5 V/CV < 0.05C & 2 s 0.5C → 3.5 V	1
Charge Discharge	1C → 5 V/CV < 0.05C & 2 s 1C → 3.5 V	3

***Table S2**. Components of the positive electrodes and upper and lower limits defined for the optimization of the slurry.*

Component	Weight fraction, wt%	
	Lower limit	**Upper limit**
LNMO	89	91
Carbon black	4	6
CMC	2	3
Binder	2	3

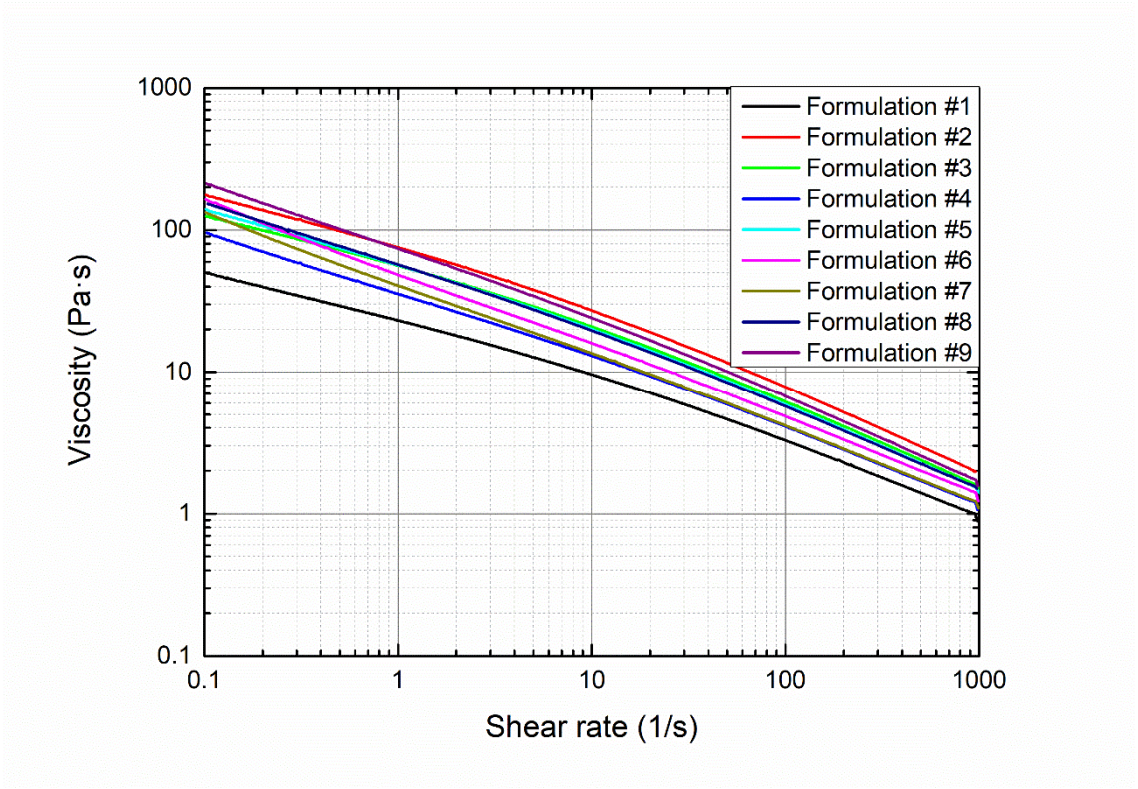

Figure S2*. Rheological studies with the different formulations: viscosity vs. shear rate curves.*

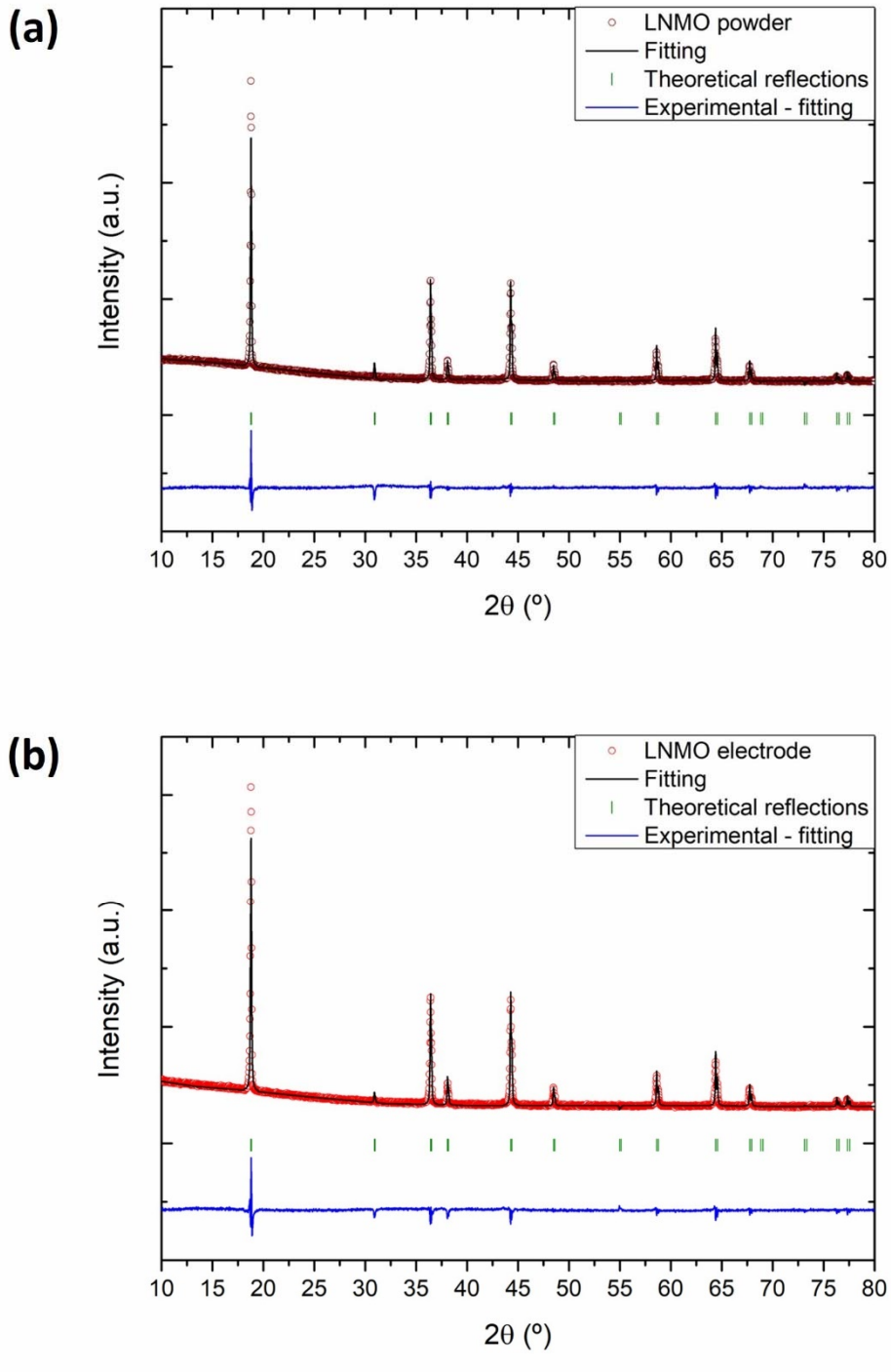

Figure S3*. X-ray diffraction patterns of the (a) LNMO powder and (b) a LNMO electrode. Patterns were fitted to LNMO phase using FULLPROF software and its theoretical reflections and the difference between the experimental and the fitting curves are included.*

LNMO powder and LNMO electrodes were subjected to XRD analysis. The patterns obtained were fitted using cation-disordered structure with face-centered cubic symmetry (space group F d -3 m). a = 8.17731 and 8.17666 Å unit cell parameters were obtained for the LNMO powder and electrode, respectively, evidencing negligible influence of the processing of the material on its crystal structure.

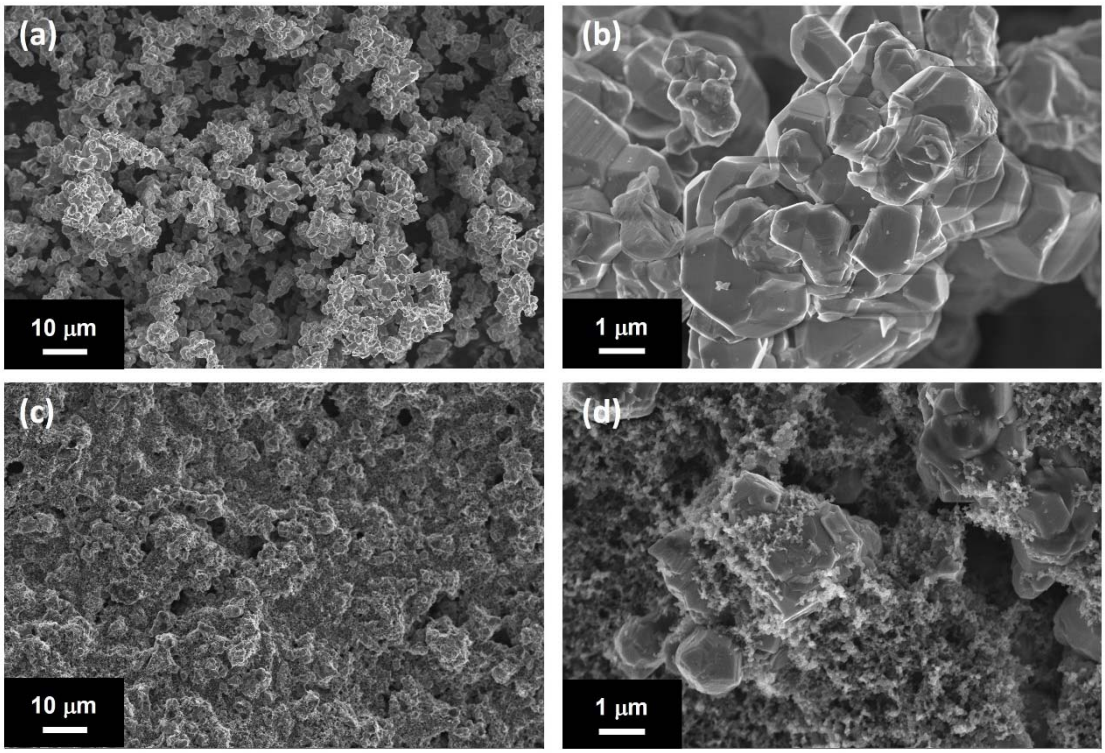

Figure S4*. FE-SEM images of the LNMO powder and a LNMO electrode: LNMO powder with magnification (a) X1000 and (b) X10000 and LNMO electrode with magnification (c) X1000 and (d) X10000.*

LNMO powder consists of 1-5 μm particle-size clusters (**Figures S4a** and **S4b**). After processing, carbon black appears covering all the electrode surface (**Figure S4c**), generating an electron-conductive matrix to overcome the insulating nature of LNMO. Nevertheless, a closer look to the electrodes (**Figure S4d**) showed that the original LNMO morphology is kept behind the carbon black matrix. No agglomerates were found in the SEM inspection.

Table S3*. Functional relations between the concentration of the electrode components and the experimental outputs.*

Equation	**R^2 (%)**
Viscosity = – 1.73[LNMO] – 78.19[C65] – 521.06[CMC] + 2.24[Binder] + 1.02[LNMO][C45] + 5.98[LNMO][CMC] + 0.14[LNMO][Binder] + 2.35[C45][CMC]	99.79
Slope = – 0.0077[LNMO] + 0.09[C65] – 0.96[CMC] – 1.09[Binder] + 0.001 [LNMO][C45] + 0.01 [LNMO][CMC] + 0.01[LNMO][Binder] – 0.02[C45][CMC]	99.57
Peel strength = 1.12[LNMO] + 94.91[C65] – 1085.85[CMC] + 438.79[Binder] – 1.34[LNMO][C45] + 11.18[LNMO][CMC] – 4.66[LNMO][Binder] + 16.95 [C45][CMC]	95.64
0.2C capacity = 1.11[LNMO] – 33.55[C65] – 185.39[CMC] + 32.48[Binder] + 0.36[LNMO][C45] + 1.95[LNMO][CMC] – 0.28[LNMO][Binder] + 3.29[C45][CMC]	39.11
2C capacity = – 0.38[LNMO] – 306.02[C65] – 487.16[CMC] + 44.40[Binder] + 3.45[LNMO][C45] + 5.09[LNMO][CMC] – 0.19[LNMO][Binder] + 10.83[C45][CMC]	90.57
1C capacity = – 0.59[LNMO] + 2.95[C65] – 449.77[CMC] – 921.56[Binder] + 0.26[LNMO][C45] – 4.60[LNMO][CMC] + 10.29[LNMO][Binder] – 5.52[C45][CMC]	98.10

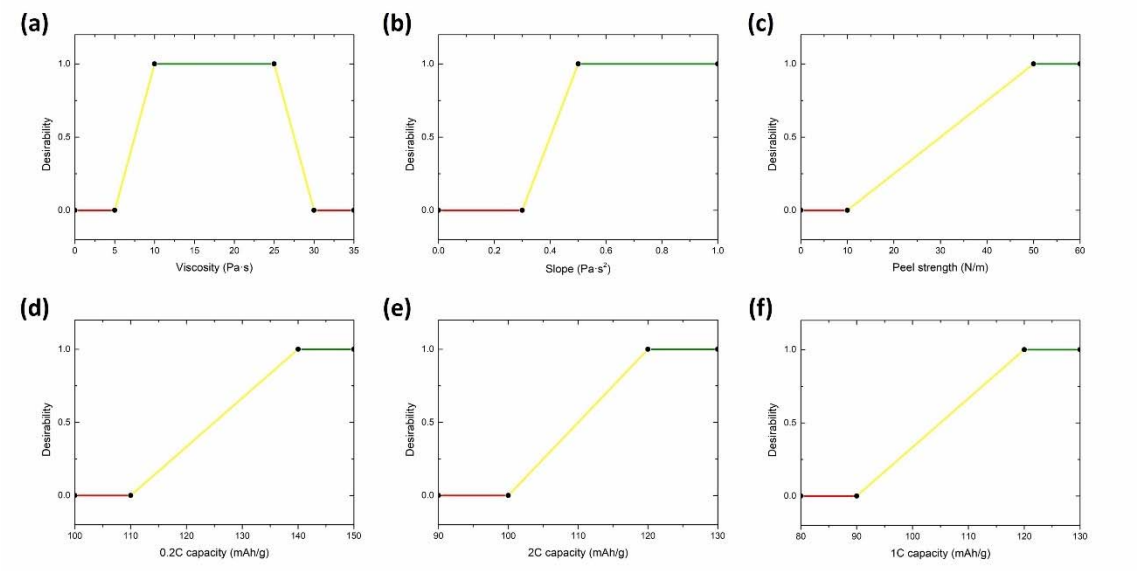

Figure S5*. Derringer-Suich desirability functions for the different experimental outputs under evaluation: (a) viscosity, (b) viscosity slope, (c) peel strength, (d) 0.2C capacity, (e) 2C capacity, and (f) 1C capacity.*

***Table S4**. Desirability of the different samples for each parameter, group of parameters and the global desirability.*

Formulation	Viscosity at 10 s^{-1}	Viscosity slope	Peel strength	Feasibility	Specific capacity at 0.2C	Specific capacity at 2C	Specific capacity at 1C	Electrochemical performance	Global desirability
1	0.920	0.204	0.670	0.774	0.609	0.823	0.821	0.891	0.928
2	0.572	0.195	0.272	0.647	0.574	0.418	0.613	0.819	0.879
3	1.000	0.105	0.185	0.580	0.601	0.516	0.591	0.834	0.873
4	1.000	0.498	0.372	0.782	0.633	1.000	0.901	0.917	0.939
5	1.000	0.342	0.561	0.801	0.553	0.604	0.246	0.761	0.878
6	1.000	0.893	0.488	0.876	0.531	0.838	0.930	0.883	0.939
7	1.000	0.794	0.864	0.950	0.6	0.855	0.394	0.830	0.926
8	1.000	0.462	0.550	0.827	0.621	0.948	0.668	0.884	0.932
9	1.000	0.639	0.522	0.852	0.643	1.000	0.810	0.910	0.946

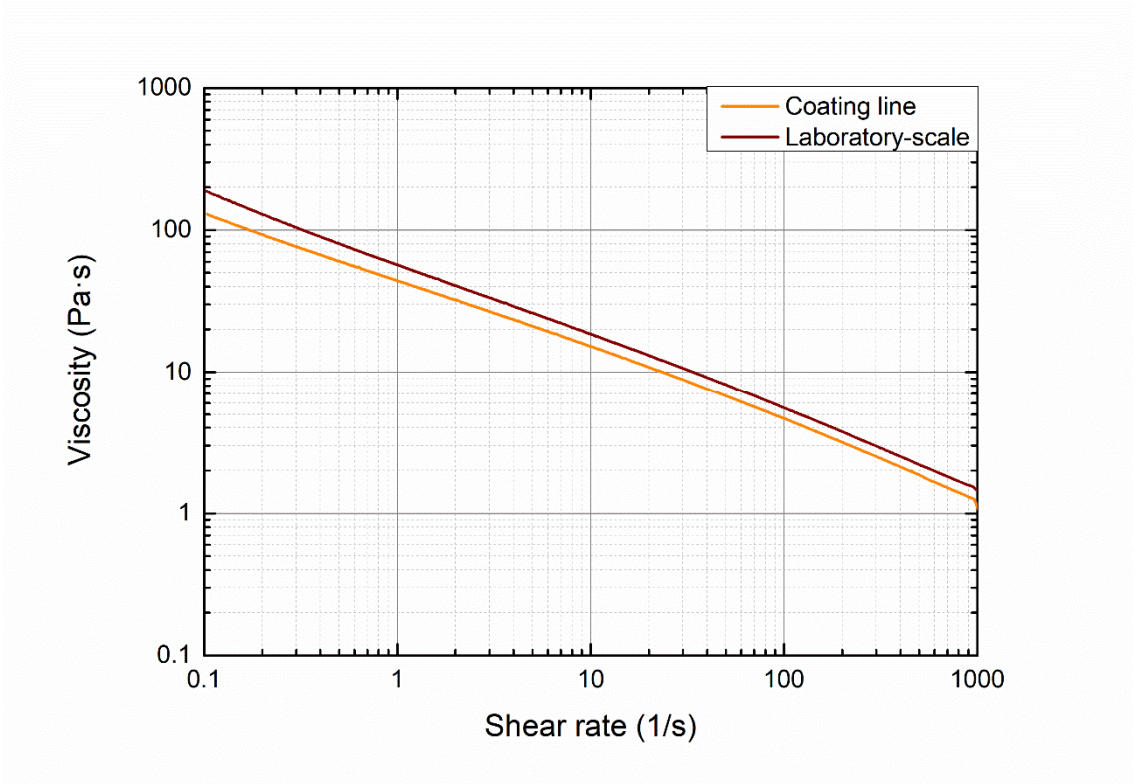

Figure S6*. Rheological analyses (viscosity vs. shear rate curves) of the laboratory-scale slurry with formulation #10 and slurry prepared for upscaling to the coating line with the same formulation.*

As expected based on the regression coefficient for equation 3 (97.83%), there are slight differences between the global desirability values obtained experimentally and the values obtained with equation 3. Both values for each formulation are compared in **Table S5**.

Table S5. *Experimental al mathematical desirability values obtained for the 9 formulations in this study.*

Formulation	Experimental D	Mathematical D
1	0.928	0.930
2	0.879	0.882
3	0.873	0.867
4	0.939	0.933
5	0.878	0.884
6	0.939	0.944
7	0.926	0.924
8	0.932	0.943
9	0.946	0.932

Interestingly, the highest experimental and mathematical desirability values are not obtained with the same formulation: the highest mathematical desirability was obtained with formulation #6. These differences are attributed to experimental errors and, thus, it is better to use the mathematical desirability as the referential parameter to evaluate the formulations.

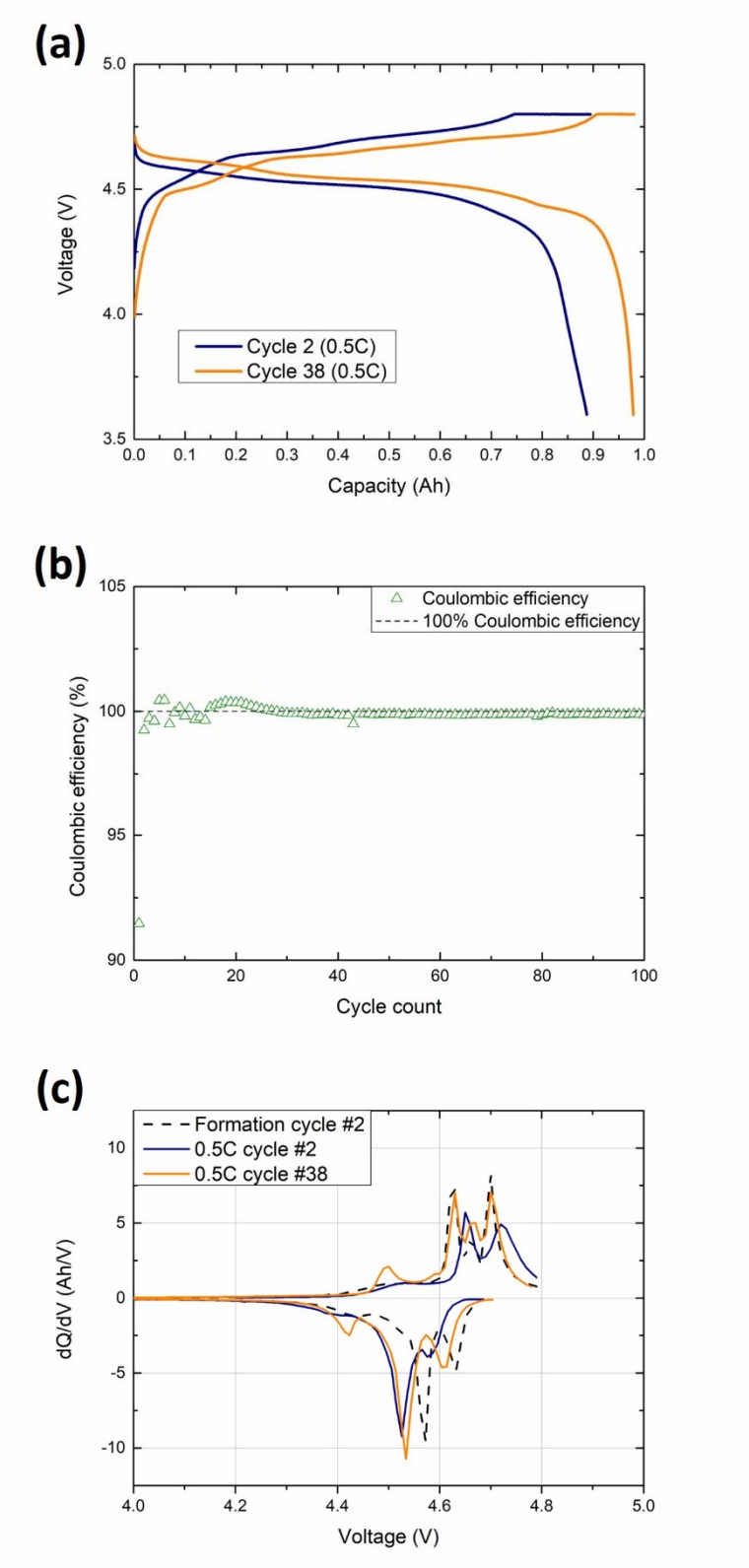

Figure S7. *(a) Voltage vs. capacity representation of the 2nd and the 38th cycles with the pouch cells cycled at 0.5C. (b) Coulombic efficiency of the cells subjected to 0.5C C-rate. (c) dQ/dV vs. voltage representation of the 2nd and the 38th cycles with the pouch cells cycled at 0.5C.*