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Supplementary information

Effect of coating on the electrochemical performance of $LiCoPO_4$ for green electrode processing

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Table S 1. Output of Rietveld Refinement of uncoated and coated LCP using Topas 4.2.

	LCP	**LCP-C1**	**LCP-C2**	**LCP-C3**	**LCP-C4**	**LCP-V**
R_{exp}	4.80	5.36	3.89	4.25	4.46	4.96
R_{wp}	5.30	5.65	4.35	4.60	4.59	5.34
R_p	4.20	4.48	3.44	3.66	3.64	4.26
GOF	1.10	1.05	1.12	1.08	1.03	1.08
DW	1.70	1.82	1.67	1.75	1.89	1.72
Refinement range (2θ): 10-80°; LP factor: 0; Order of background parameter (Chebichev polynomial): 15						
Specimen displacement (mm)	-0.243(2)	-0.154(2)	-0.232(2)	-0.297(3)	-0.339(3)	-0.127(1)
Phase 1	**$LiCoPO_4$**					
B_{eq}** ($Å^2$)	4.4(1)	5.6(1)	6.3(1)	5.8(1)	3.8(1)	5.9(1)
R-Bragg	1.861	2.164	2.544	1.674	1.206	2.914
Space group	*Pnma* (62)					
Lattice parameter						
a (Å)	10.2069(3)	10.2064(4)	10.2052(3)	10.2005(5)	10.2061(6)	10.2054(2)
b (Å)	5.9231(2)	5.9229(2)	5.9242(2)	5.9328(3)	5.9313(3)	5.9221(1)
c (Å)	4.7011(2)	4.7006(2)	4.7011(2)	4.7043(3)	4.7049(3)	4.7004(1)
Crystallite size						
Lorentzian (nm)	190(7)	127(5)	120(3)	96(3)	131(10)	210(6)
Gaussian (nm)	145(5)	109(5)	136(6)	108(6)	59(2)	279(17)
Phase 2	**Co_2P**					
B_{eq}**($Å^2$)	=B_{eq} (Phase 1)					
R-Bragg	0.310	0.607	0.514	1.139	0.746	0.296
Space group	*Pnma* (62)					
Lattice parameter						
a (Å)	5.63(7)	5.64(5)	5.652(6)	5.659(6)	5.666(5)	5.65(9)
b (Å)	3.52(4)	3.52(3)	3.511(4)	3.511(4)	3.510(3)	3.50(6)
c (Å)	6.66(8)	6.61(6)	6.610(7)	6.609(7)	6.611(6)	6.6(1)
Crystallite size						
Lorentzian (nm)*	50					
Gaussian (nm)*	50					
Phase 1 (%)	99.7(2)	99.6(2)	97.4(2)	96.6(2)	95.9(2)	99.8(2)
Phase 2 (%)	0.3(2)	0.4(2)	2.6(2)	3.4(2)	4.1(2)	0.2(2)

*fixed parameter

**B_{eq} was simultaneously refined for all atoms set to the same value of B_{eq}

The initial structural models used for the Rietveld refinement were taken from the data of F. Kubel, Zeitschrift für Kristallographie - Crystalline Materials 209 (1994), 755 (ICSD 400625) for phase 1 ($LiCoPO_4$) and S. Rundqvist, Acta Chem. Scan, 14 (1960) 1961-1979 (ICSD 43685) for phase 2 (Co_2P).

Table S2. Results of impedance spectroscopy fitting for uncoated and coated LCP electrodes at 4.8 V of discharge during the 112th cycle using ZView 2 applying an equivalent circuit model.

	LCP	**LCP/C1**	**LCP/C2**	**LCP/V**
112th cycle				
χ^2	0.0064398	0.009176	0.007447	0.000409
R_S	9.80	7.99	7.04	2.286
R_S (error)	0.329	0.465		0.066
R_1	26.93	10.54	10.56	23.31
R_1 (error)	0.558	0.583	0.165	5.574
CPE_1-T	5.5E-06	5.0E-06	4.0E-06	3.9E-05
CPE_1-T (error)	0.021	0.046	0.021	0.028
CPE_1-P	0.74	0.75	0.81	0.66
CPE_1-P (error)	2.8	6.1	2.5	4.2
R_2				241.4
R_2 (error)				4.9
CPE_2-T				0.00011
CPE_2-T (error)				0.000008
CPE_2-P				0.59
CPE_2-P (error)				0.019
CPE_W-T	0.005	0.007	0.009	0.0016
CPE_W-T (error)	0.000048	0.000067	0.000087	0.000017
CPE_W-P	0.59	0.53	0.57	0.45
CPE_W-P (error)	0.004	0.004	0 004	0.004

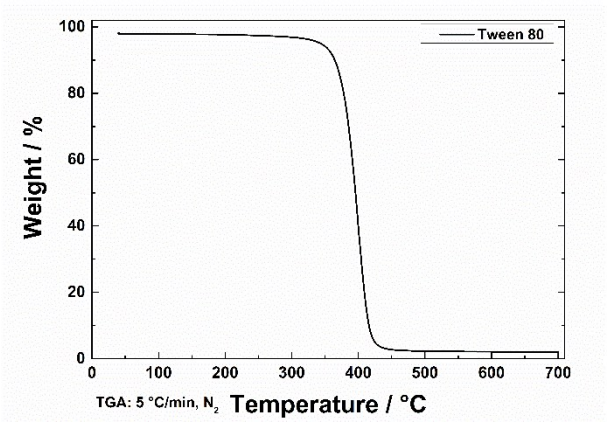

Figure S1. TGA of Tween 80® in N_2 atmosphere.

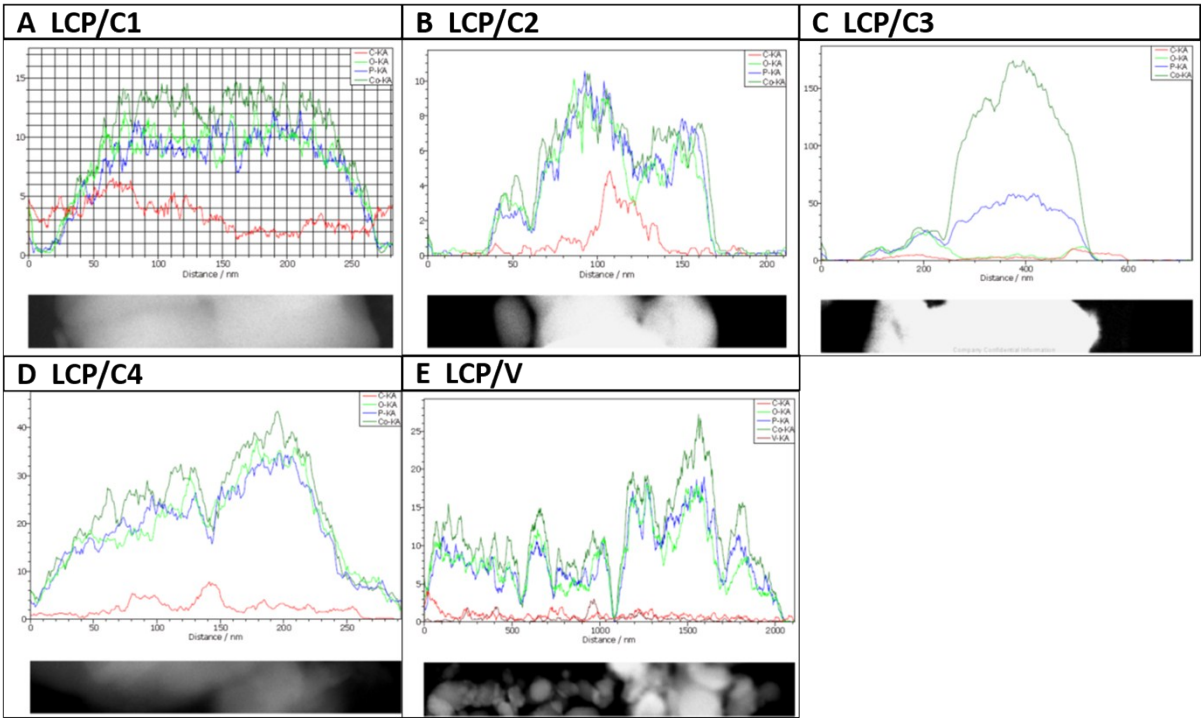

Figure S2. EDX line scan of LCP with different coatings: A LCP/C1 (sucrose), B LCP/C2 (sucrose), C LCP/C3 (PAA), D LCP/C4 (Tween 80), E LCP/V (V_2O_5).

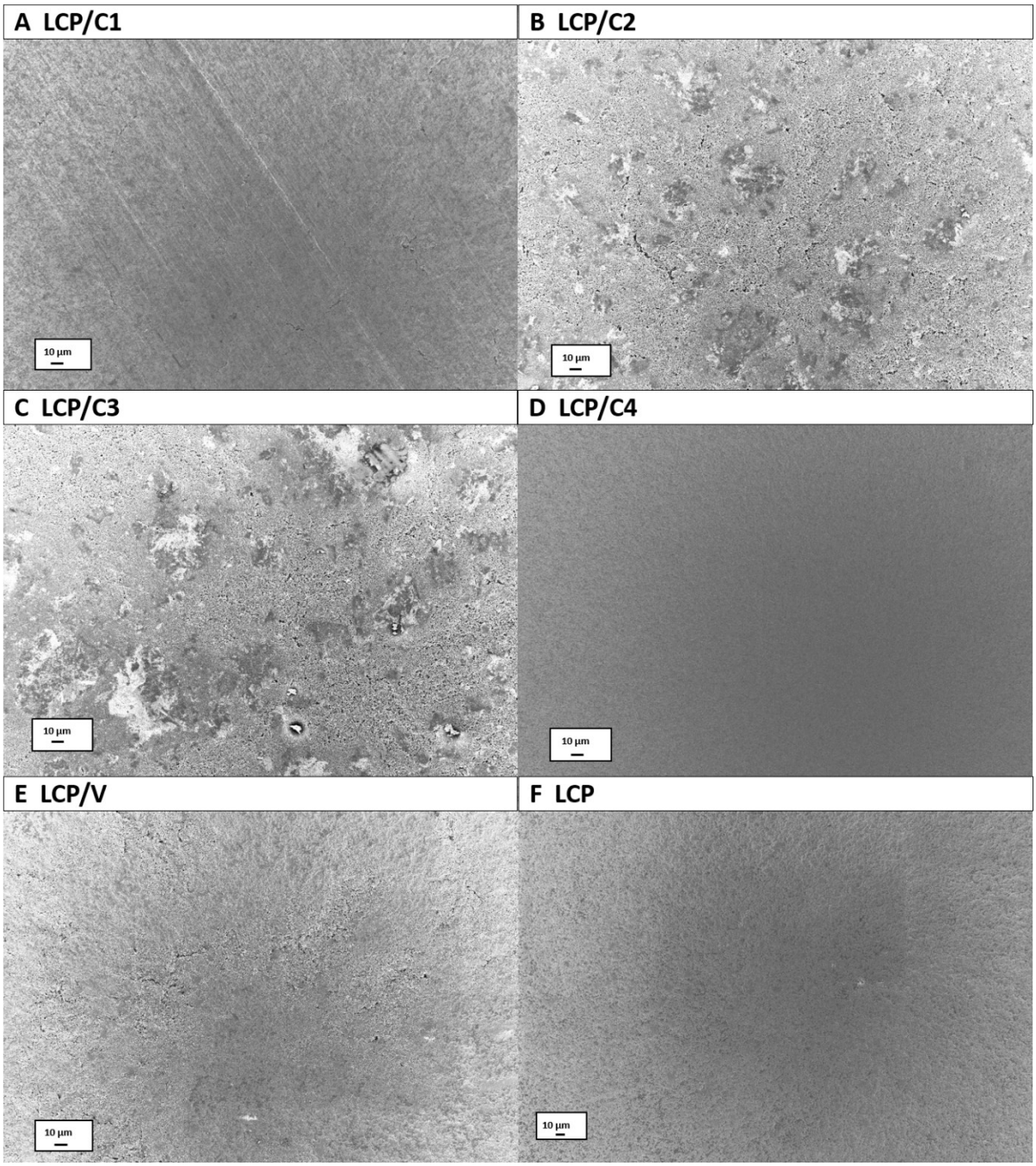

Figure S3. SEM images of electrodes with pristine and coated LCP A: 20 wt.% sucrose, B: 10 wt.% sucrose, C: 20 wt.% polyacrylic acid, D: 40 wt.% Tween 80®, E: 3 wt.% V_2O_5, F: pristine LCP.

The morphology of pristine and coated LCP electrodes made using the aqueous binder (CMC), subsequently pressed to reduce the porosity was also investigated by SEM (Figure S3). Pristine LCP, LCP/C1 and LCP/C4 (A, D, F) showed very smooth and homogeneous electrode surfaces. On the other hand, some agglomerates and cracks were detected for LCP/C2 (B) while LCP/C3 (C) showed a very inhomogeneous distribution of active material and conductive carbon along with a large amount of agglomerates. LCP/V (E) showed some cracks, but an overall smooth electrode surface and no major inhomogeneities due to active material particles' agglomeration.[14]

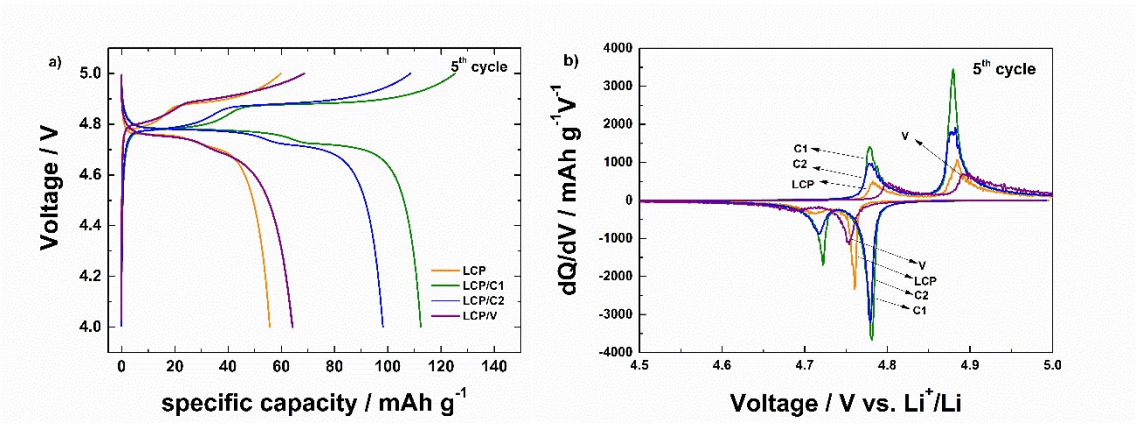

Figure S4. a) Voltage profile and b) differential capacity plot of uncoated and coated LCP (selected materials) during the 5th cycle (C/10).