Supporting information

Effect of Nanostructured and Open-Porous Particle Morphology on Electrode Processing and Electrochemical Performance of Li-Ion Batteries

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Structural analysis (XRD)

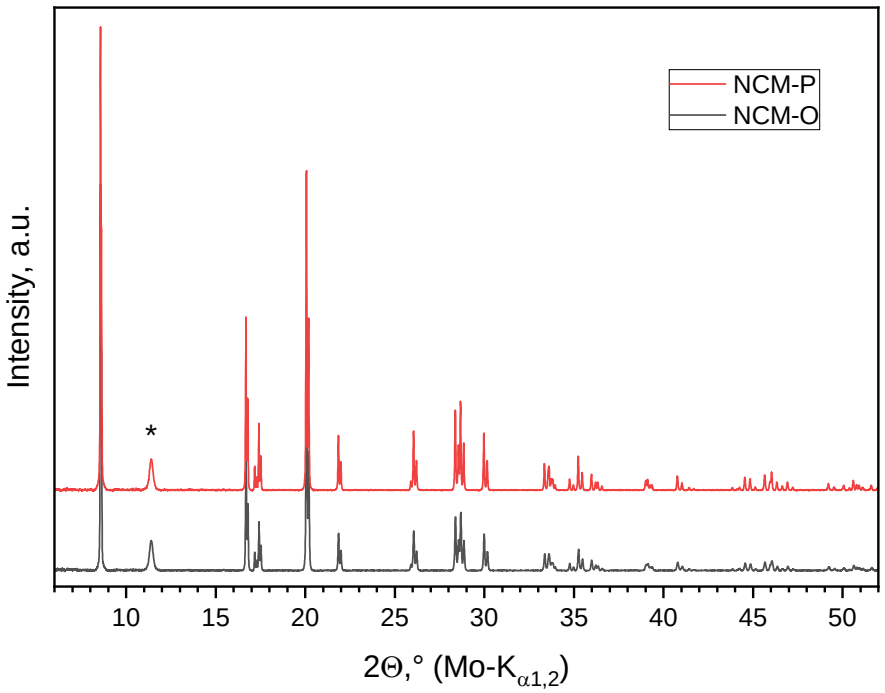

Figure S1: XRD patterns of the original Toda NM3100 (NCM-O) and the processed NCM-P (850°C, 5h); * reflex belongs to the used glass capillary tube.

Powder X-ray diffraction (XRD) was performed using a laboratory diffractometer optimized for battery research (Figure S1). The diffractometer is equipped with a molybdenum microfocus rotating anode (Mo−Kα1,2).

Materials Characterization ICP-OES

Elemental analysis was performed at IAM-AWP (chemical analysis), KIT (Table S1). The composition of the samples was measured three times. Li, Ni, Co, Mn was analyzed by ICP-OES, O by carrier gas hot extraction.

Table S1: Elemental analysis of NCM-O and NCM-P by ICP-OES

	NCM-O				NCM-P, 850°C, 5h			
	Mass%			Atom%	Mass%			Atom%
	AV	SD	±	AV	AV	SD	±	AV
Li	7,08	0,02	0,15	**24,9**	7,02	0,03	0,15	**24,9**
O	32,8	0,3	2,6	**50,0**	32,5	0,7	2,6	**50,1**
Mn	18,87	0,07	0,32	**8,38**	18,68	0,05	0,32	**8,38**
Co	20,09	0,05	0,38	**8,32**	19,84	0,03	0,38	**8,30**
Ni	20,07	0,03	0,26	**8,35**	19,82	0,03	0,26	**8,32**

AV: average

SD: standard deviation

±: measurement uncertainty

Adhesion Tests – Delamination Behavior

Inspection of the stripped current collector foil following the adhesion strength measurements revealed predominantly adhesive failure between electrode layer and aluminum foil (Figure S2).

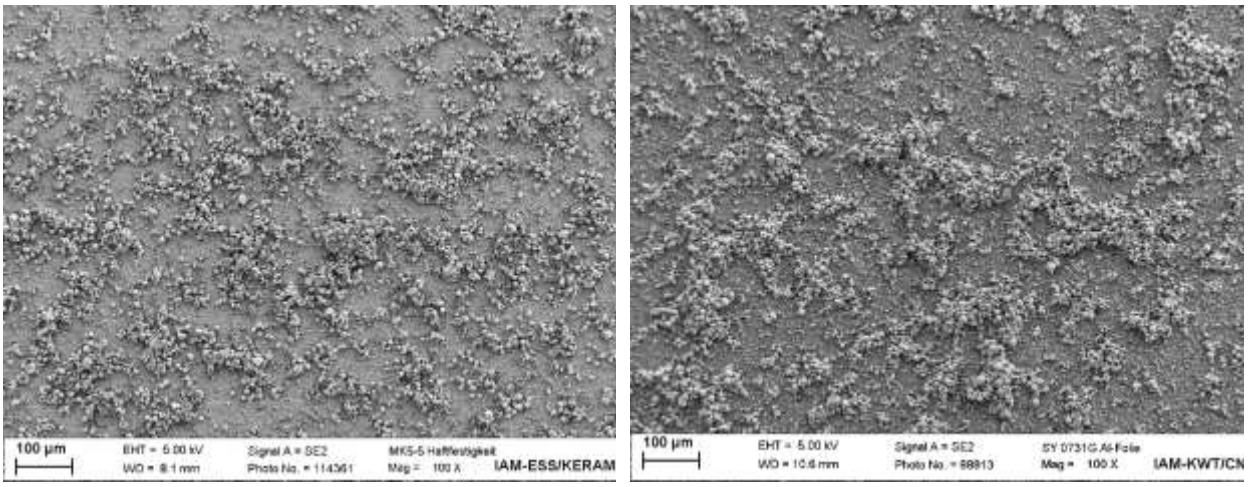

Figure S2: SEM micrographs of electrode layer residues adhering to current collector after adhesion tests; left = NCM-O 444, right = NCM-P 444.

EDS-Analysis of particle cross-sections

Measurements were performed using an Ultim Extreme silicon drift detector from Oxford Instruments with AZtec software (version 4.2). The selected acceleration voltage of 4kV is relatively low, but chosen as a balance between high spatial resolution in combination with reduced sample damage on the one hand and sufficient count rate on the other hand. From the marked areas, which cover secondary particle cross-sections of NCM-P 444 (Figure S3) and NCM-O 444 (Figure S4), spectra were captured to determine the F content as an indicator for the presence (or absence) of PVDF binder. The NCM-P 444 sample shows clearly fluorine and carbon in the porous network (Table S2). However, due to a peak overlap of fluorine-(K-line) and Mn and Co L-lines it is likely that to a certain degree the F content is overestimated. For that reason, we also measured cross-sections of the original material without open porosity where PVDF binder cannot be expected (Table S3). The detection of 0.9 at% (NMC-O_444) is therefore attributed to the before mentioned peak-overlap. Hence, the substantially higher amount of F inside the NCM-P particles indicates the presence of PVDF binder.

The amounts of the other elements are presented primarily to have a reference for the C and F concentration. EDS-analysis delivers under these circumstances only semi-quantitative results and is not comparable to ICP-OES measurements (note that it is not possible to detect Li in NMC).

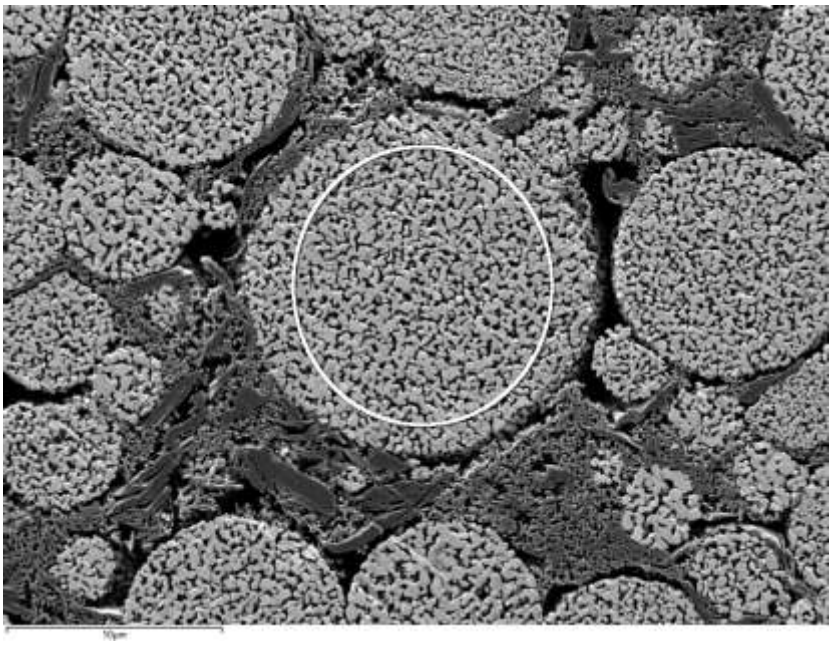

Figure S3: Area of analysis of cross-sectioned NCM-P_444 electrode

Table S2: EDS analysis of NCM-P particle

Element	Line Type	Wt%	Wt% Sigma	Atomic%	Standard Label
C	K series	2.29	0.14	**5.53**	Pure Element
O	K series	32.22	0.2	58.26	SiO_2
F	K series	3.04	0.09	**4.63**	CaF_2
Mn	L series	25.1	0.33	13.22	Pure Element
Co	L series	21.2	0.27	10.41	Pure Element
Ni	L series	16.14	0.19	7.95	Pure Element
Total:		100		100	

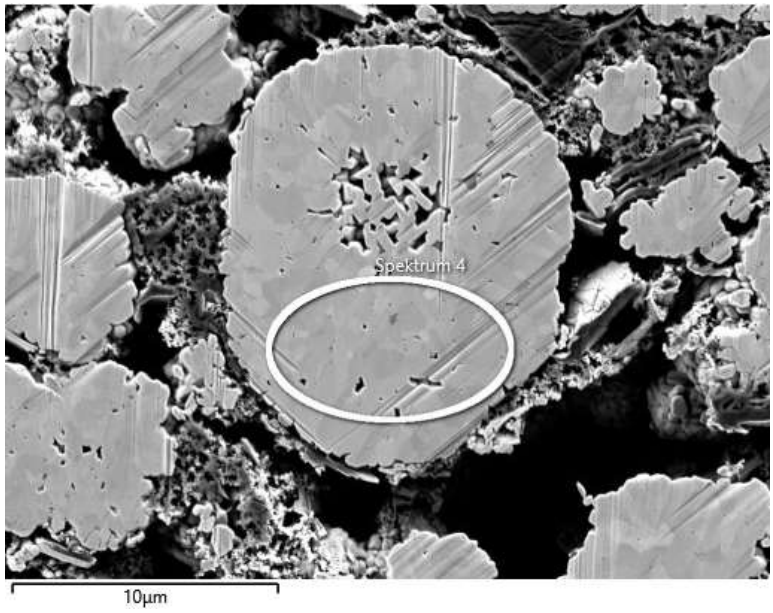

Figure S4: Area of analysis of cross-sectioned NCM-O 444 electrode

Table S3: EDS analysis of pristine NCM-O particle

Element	Line Type	Wt%	Wt% Sigma	Atomic %	Standard Label
C	K series	1.02	0.12	2.73	Pure Element
O	K series	28.32	0.15	56.98	SiO2
F	K series	0.55	0.06	0.92	CaF2
Mn	L series	26.04	0.27	15.26	Pure Element
Co	L series	23.73	0.22	12.96	Pure Element
Ni	L series	20.34	0.17	11.15	Pure Element
Total:		100		100	