

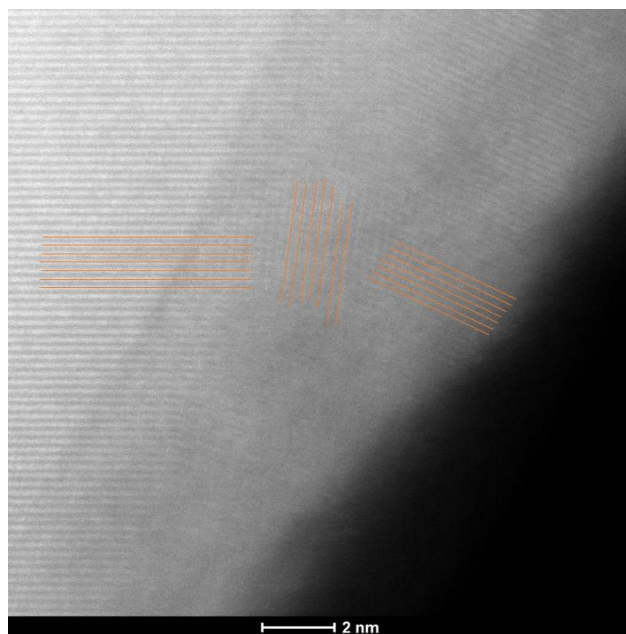
# Study of Immersion of $\text{LiNi}_{0.5}\text{Mn}_{0.3}\text{Co}_{0.2}\text{O}_2$ Material in Water for Aqueous Processing of Positive Electrode for Li-Ion Batteries

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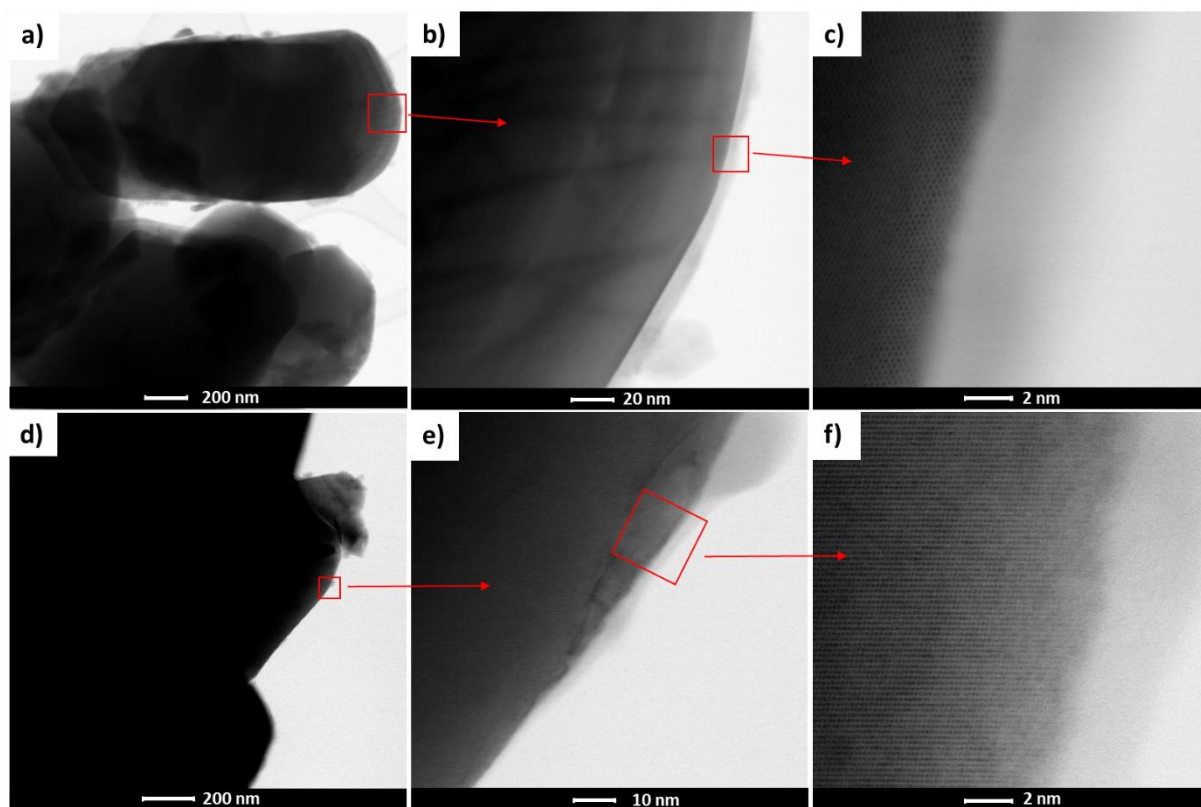
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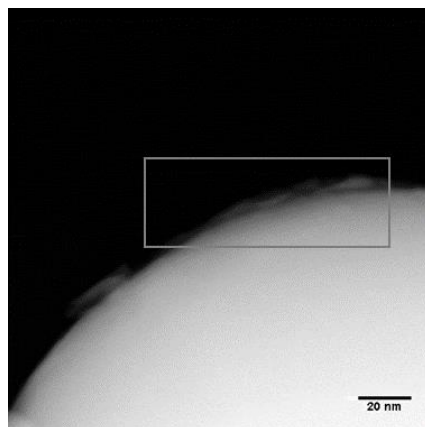
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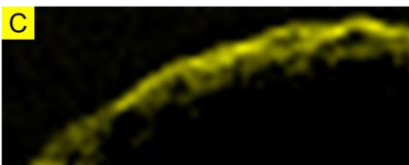


**Figure S1:** HRSTEM-HAADF image of NMC532 powder recovered after 1 h immersion in acidified water followed by water evaporation at 50 °C (sample P2). The lines indicate different orientations of the TM layers.

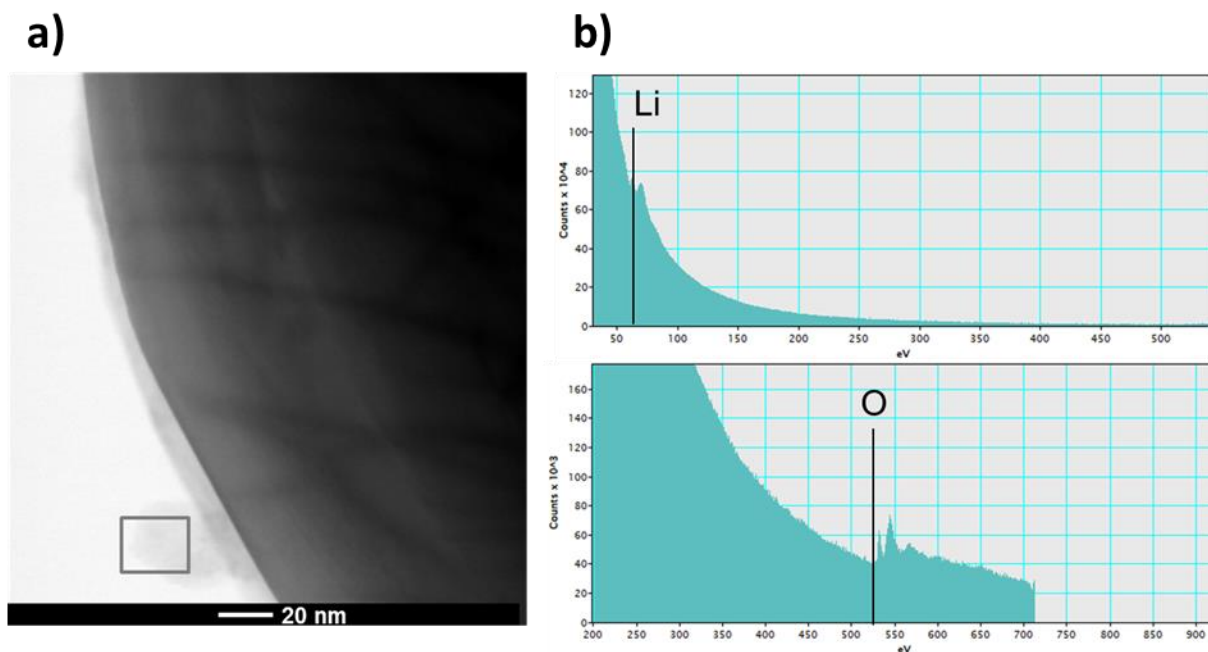


**Figure S2:** STEM-BF images of NMC532 powders recovered by water evaporation at 50 °C after 1 h immersion in (a) water (sample P1) and (d) acidified water (sample P2). (c) and (f) correspond to the selected area in (b) and (e), which are themselves extracted from images (a) and (d), respectively.

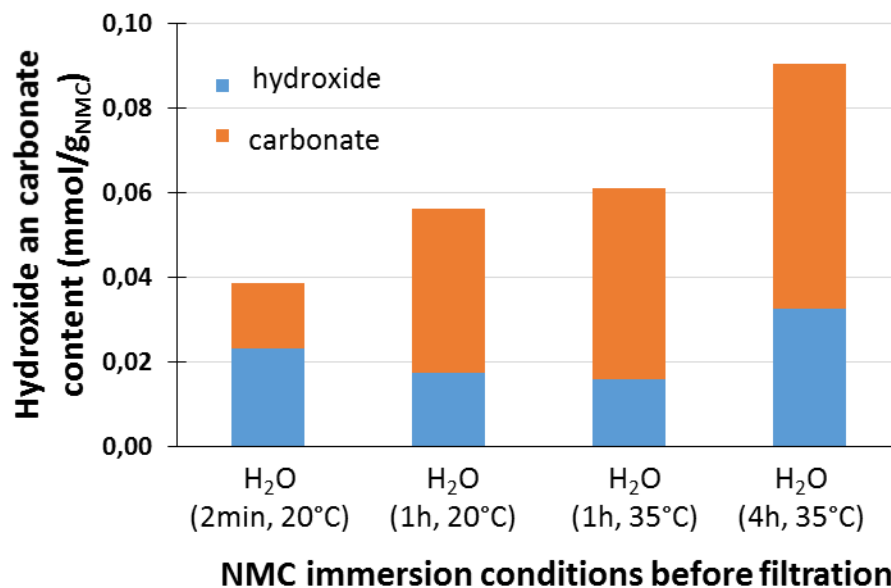




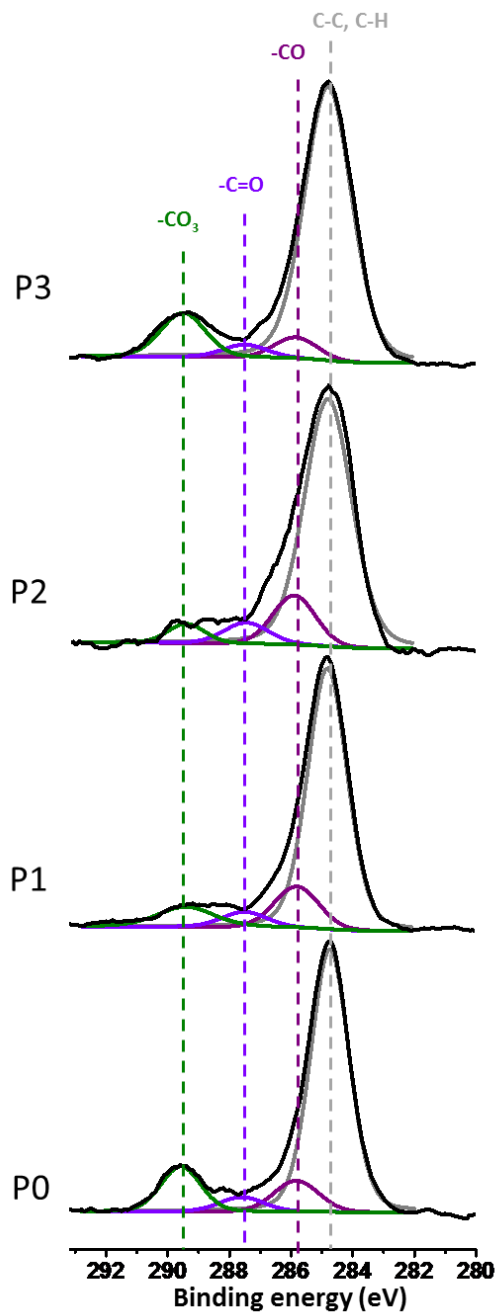
**Figure S3:** STEM-EELS analysis of NMC532 powder after immersion in acidified water (sample P2). The carbon mapping on the lower magnification image evidences the presence of carbon in the surface layer.



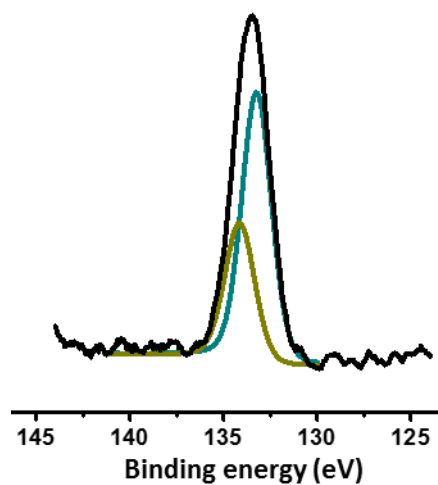
**Figure S4:** (a) BF-STEM image of NMC532 powder after immersion in water (sample P1). (b) EELS analysis of the zone marked on **Figure S4a** shows that it is composed of LiOH, as the same amount of Li and O was obtained from the elemental quantification.



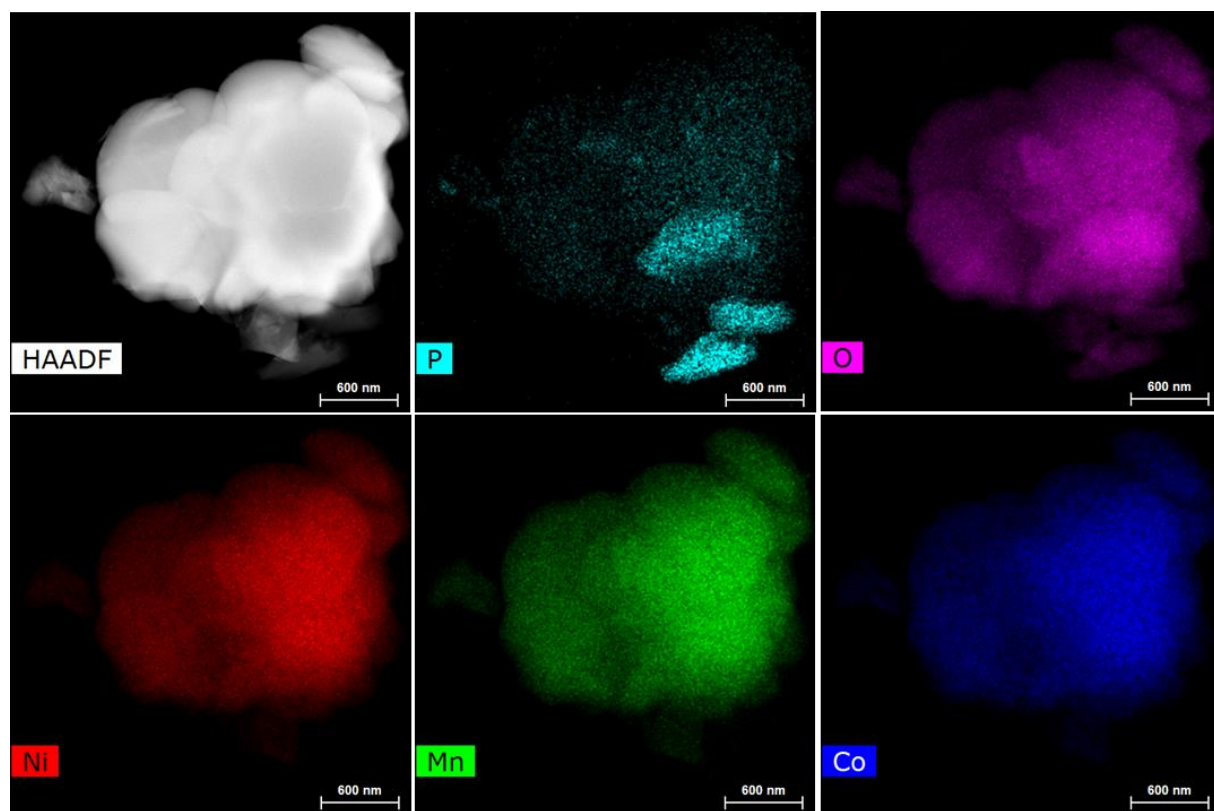
**Figure S5:** Hydroxide and carbonate content measured by pH titration on aqueous filtrates. The filtrates were obtained after filtration of NMC532 powder immersed in water at ambient temperature (20 °C) for 2 min or 1 h, or immersed in water heated at 35 °C for 1 h or 4 h. The amount of hydroxides and carbonates is increased when the immersion duration and temperature increase.



**Figure S6:** C 1s XPS spectra of different NMC532 samples: pristine (P0), soaked 1 h in water or acidified water and recovered after water evaporation (P1 and P2), and stored 1 year in air (P3).

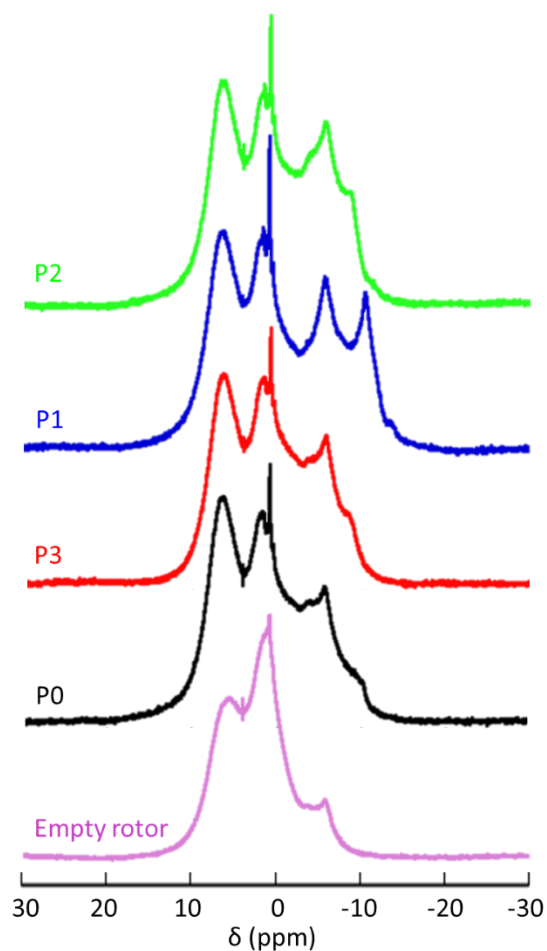


**Figure S7:** P 2p XPS spectrum of sample P2. This peak envelope centered at 133.5 eV can be deconvoluted with the overlapping shapes of P 2p<sub>3/2</sub> and P 2p<sub>1/2</sub> spaced with 0.9 eV. It is ascribed to phosphate.

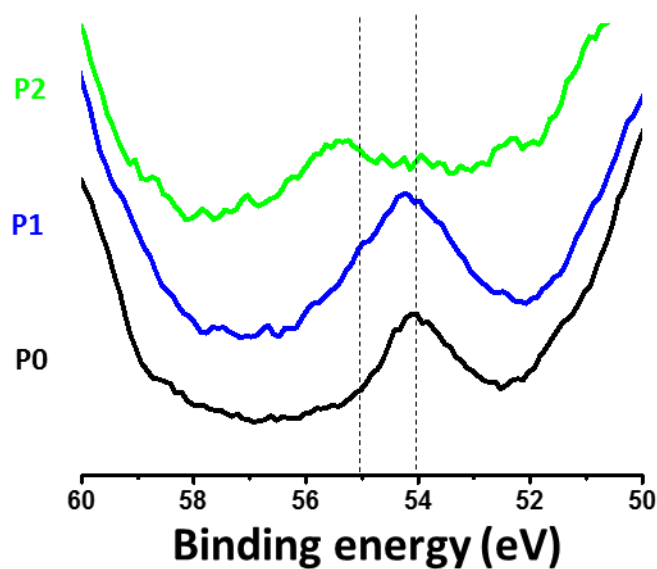


**Figure S8:** TEM-HAADF image and EDX mapping of elements Ni, Mn, Co, O, and P of a particle from sample P2.





**Figure S9:**  $^1\text{H}$  NMR spectra of the powders P0, P1, P2, and P3 normalized with the scan number, receiver gain and sample mass. The spectrum recorded with the empty rotor is also displayed. All measurements were carried out on a spectrometer 200 MHz at 25 kHz. The samples P0, P2, and P3 have a similar signal, with peaks positioned at the same chemical shift as the empty rotor signal. The sample P1 displays an additional strong peak at -10.5 ppm which could be attributed to protons inserted into the NMC bulk.

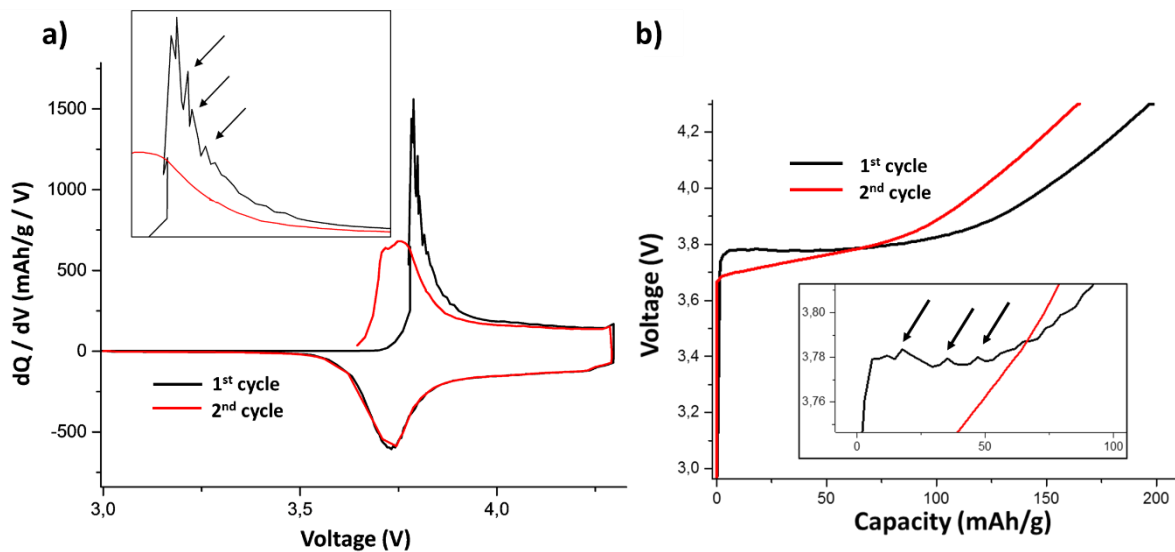


**Figure S10:** Li 1s spectra of powders P0, P1, and P2. The contribution near 54.0 eV is ascribed to lithium in the NMC bulk, while the contribution near 55.0 eV is attributed to surface species (LiOH, Li<sub>2</sub>CO<sub>3</sub>).

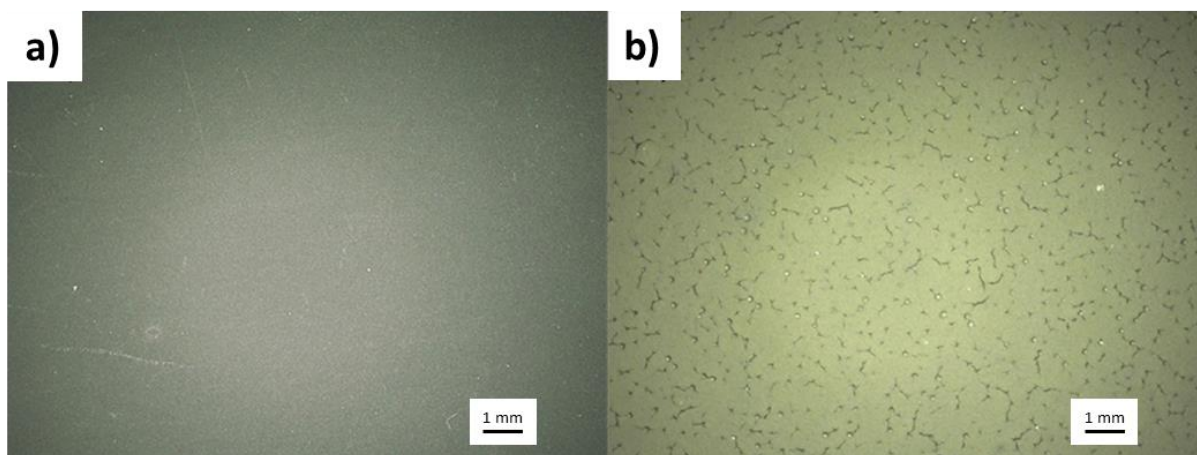
**Table S1:** Charge and discharge capacities of the first and second cycle at 0.1 C of electrodes P0<sub>NMP</sub>, P0<sub>H<sub>2</sub>O</sub> and P0<sub>H<sub>3</sub>PO<sub>4</sub></sub>, in half and full cell configuration. These results were repeated over several cells that had on average the same capacities as shown here.

Half-cells	P0 <sub>NMP</sub>	P0 <sub>H<sub>2</sub>O</sub>	P0 <sub>H<sub>3</sub>PO<sub>4</sub></sub>
1 <sup>st</sup> cycle	$Q_{ch} = 192 \text{ mAh/g}$ $Q_{dis} = 167 \text{ mAh/g}$	$Q_{ch} = 188 \text{ mAh/g}$ $Q_{dis} = 165 \text{ mAh/g}$	$Q_{ch} = 185 \text{ mAh/g}$ $Q_{dis} = 164 \text{ mAh/g}$
2 <sup>nd</sup> cycle	$Q_{ch} = 167 \text{ mAh/g}$ $Q_{dis} = 166 \text{ mAh/g}$	$Q_{ch} = 164 \text{ mAh/g}$ $Q_{dis} = 164 \text{ mAh/g}$	$Q_{ch} = 160 \text{ mAh/g}$ $Q_{dis} = 163 \text{ mAh/g}$
Full-cells	P0 <sub>NMP</sub>	P0 <sub>H<sub>2</sub>O</sub>	P0 <sub>H<sub>3</sub>PO<sub>4</sub></sub>
1 <sup>st</sup> cycle	$Q_{ch} = 190 \text{ mAh/g}$ $Q_{dis} = 162 \text{ mAh/g}$	$Q_{ch} = 188 \text{ mAh/g}$ $Q_{dis} = 158 \text{ mAh/g}$	$Q_{ch} = 184 \text{ mAh/g}$ $Q_{dis} = 155 \text{ mAh/g}$

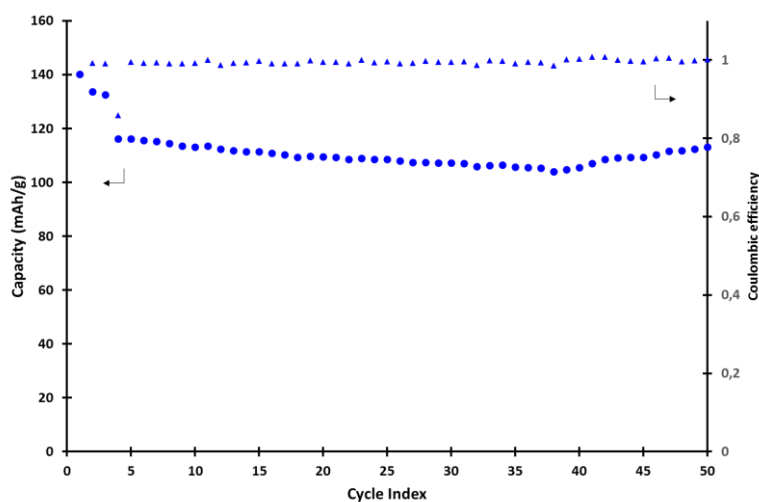
$2^{nd}$ cycle	$Q_{ch} = 165 \text{ mAh/g}$ $Q_{dis} = 160 \text{ mAh/g}$	$Q_{ch} = 161 \text{ mAh/g}$ $Q_{dis} = 157 \text{ mAh/g}$	$Q_{ch} = 160 \text{ mAh/g}$ $Q_{dis} = 154 \text{ mAh/g}$
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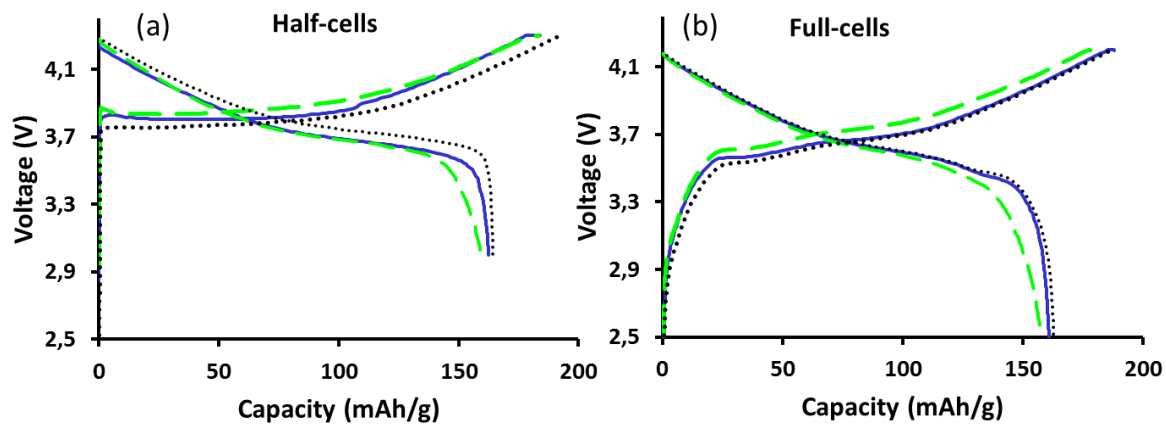
**Figure S11:** (a) Differential capacity plot at 0.1 C rate of electrode P0<sub>H2O</sub> in half-cell configuration, and (b) corresponding voltage profile. The spikes pointed by the arrows may be due to the formation of gas bubbles. As gas bubbles are formed at the electrode surface, the available electroactive surface is reduced, causing voltage rise and drop when the bubble leaves the surface.



**Figure S12:** Image of the surface of unpressed electrodes (a)  $\text{P0}_{\text{H}_3\text{PO}_4}$  and (b)  $\text{P0}_{\text{H}_2\text{O}}$ . The cracks visible on the surface of  $\text{P0}_{\text{H}_2\text{O}}$  are caused by  $\text{H}_2$  release during drying of the electrode coating, owing to the corrosion of the aluminum collector.



**Figure S13:**  $\text{P0}_{\text{H}_2\text{O}}$  cycled against a fresh Li anode. The cell is discharged at 0.1 C rate for the first three cycles, then the discharge current is increased at 0.5 C rate. The cathode is able to deliver close to 115 mAh/g, which confirms that the end of life of the cell containing  $\text{P0}_{\text{H}_2\text{O}}$  in **Figure 7a** was due to the lithium anode.



**Figure S14 :** Voltage profile of the first cycle at 0.1 C rate of electrodes P0<sub>NMP</sub> (black dotted line), P1<sub>NMP</sub> (blue solid line) and P2<sub>NMP</sub> (green dashed line) in (a) half-cell configuration and (b) full cell configuration.