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Supplementary information

Activated Nanolithia as an Effective Prelithiation Additive for Lithium-Ion Batteries

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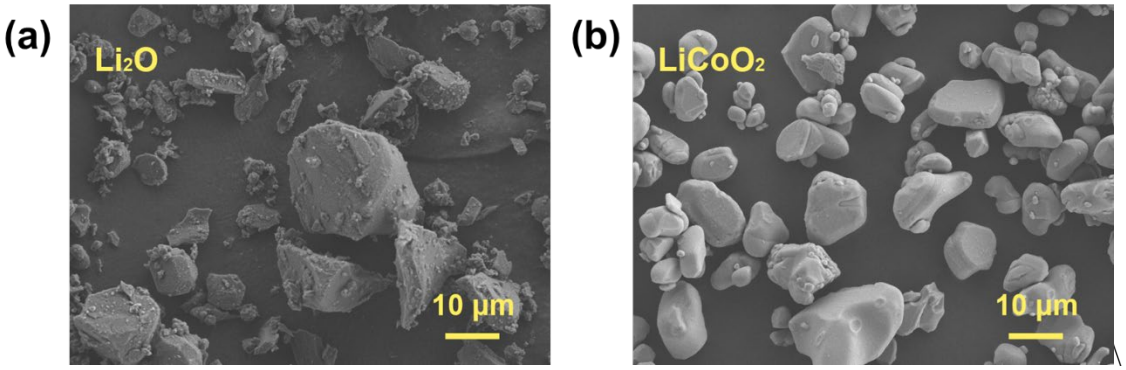

Figure. 1 SEM images of commercial Li_2O powder (a) and commercial $LiCoO_2$ powder (b).

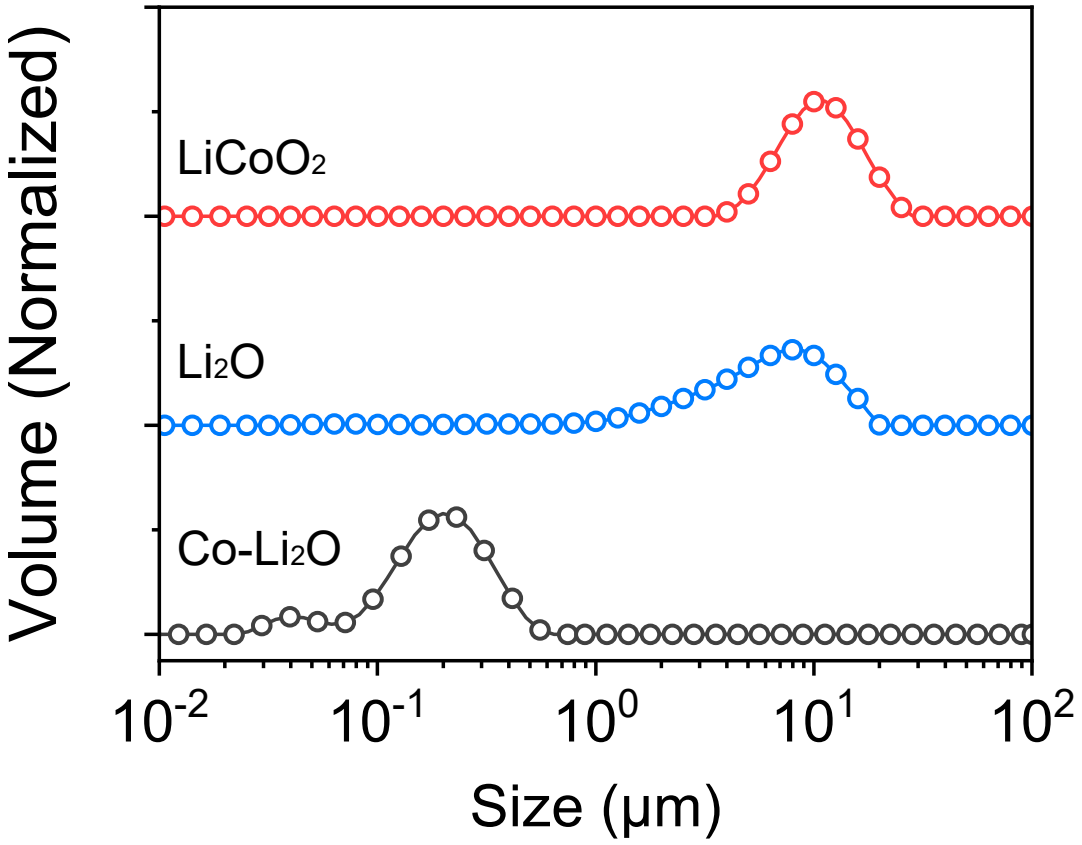

Figure. 2 Particle size distribution of $LiCoO_2$, Li_2O and Co-Li_2O.

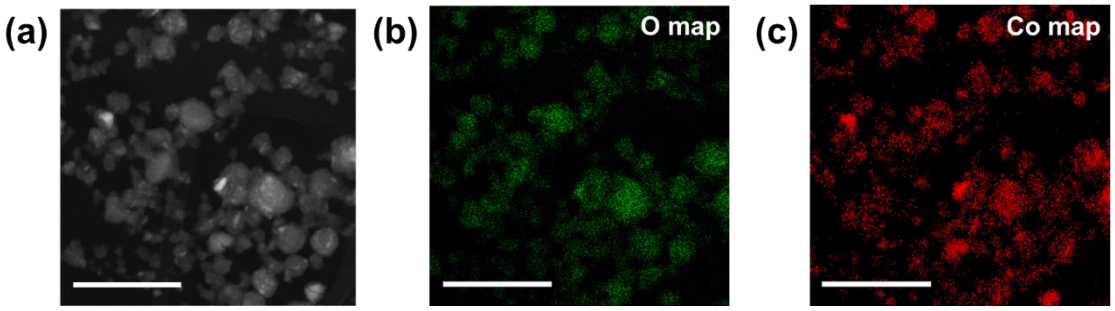

Figure. 3 (a) TEM image of Co-Li_2O. (b, c) EDS mapping of Co-Li_2O. Scale bar: 1 μm.

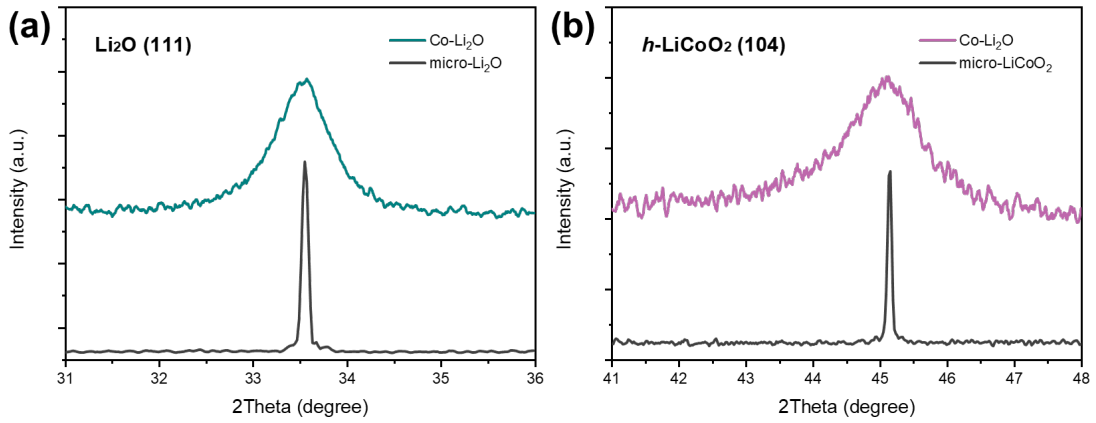

Figure. 4 XRD peaks broadening of Co-Li_2O.

The crystallite sizes can be calculated by Sherrer's equation:

$$D_{hkl}= \frac{0.89\lambda}{\beta_{hkl}\cos\theta}$$

Where D_{hkl} is the thickness of the primary particle in [*hkl*] direction (representing the diameter of the primary particle), λ is the wavelength of the incident X-ray, β_{hkl} is the full width at half maximum (FWHM) and θ is the diffraction angle of (*hkl*). The results are listed in Supplementary Table 1.

Table. 1 Stimulated peaks information and calculated crystallite sizes.

Peak	Instrumental FWHM (°)	FWHM (°)	2Theta (°)	Crystallite size (nm)
Li_2O (111)	0.06309	0.78331	33.52049	11
$LiCoO_2$ (104)	0.06988	1.53069	45.0571	6

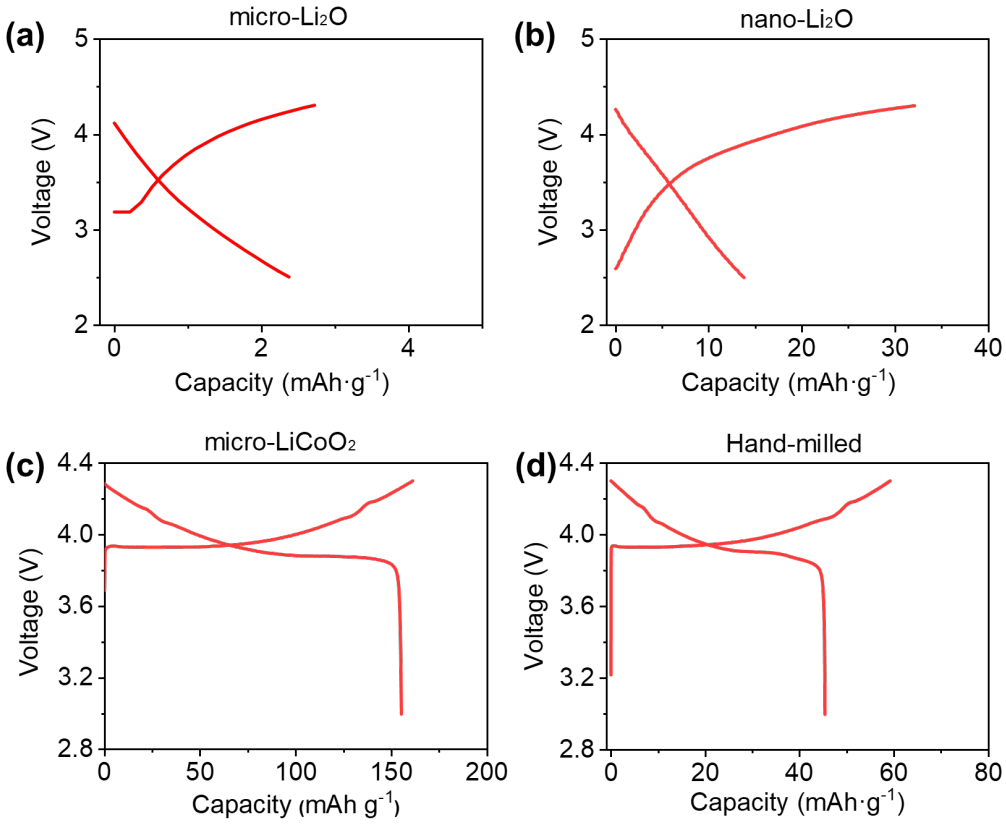

Figure. 5 Initial charge-discharge voltage profile of (a) micro-Li_2O, (b) nano-Li_2O, (c) micro-$LiCoO_2$ and (d) Hand-milled Li_2O-$LiCoO_2$ composite.

Table. 2 Calculation of extracted Li ratio in Li_2O.

x (Li_2O)	Theoretical irreversible capacity (mAh g^{-1})	Experimental irreversible capacity (mAh g^{-1})	Extracted Li ratio (%)
1 (pure Li_2O)	1793.9	18.9	1
0.9	1614.5	806.5	50
0.8	1435.1	1001.5	70
0.7	1255.7	1211.9	97
0.6	1076.3	1110.1	103
0.5	897.0	937.5	105
0.4	717.6	701.7	98
0 (pure $LiCoO_2$)	0	5.9	-

The capacity from $LiCoO_2$ is assumed to be fully reversible, while the capacity from Li_2O is assumed to be fully irreversible. Thus the extracted Li ratio in Li_2O is calculated by

$$\text{Extracted Li ratio} = \frac{\text{Experimental irreversible capacity}}{\text{Theoretical irreversible capacity}}$$

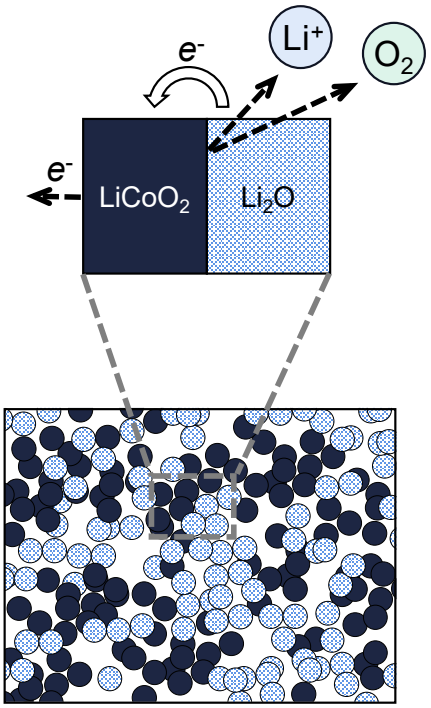

Figure. 6 Schematic diagram of the origin of facile Li_2O decomposition kinetic.

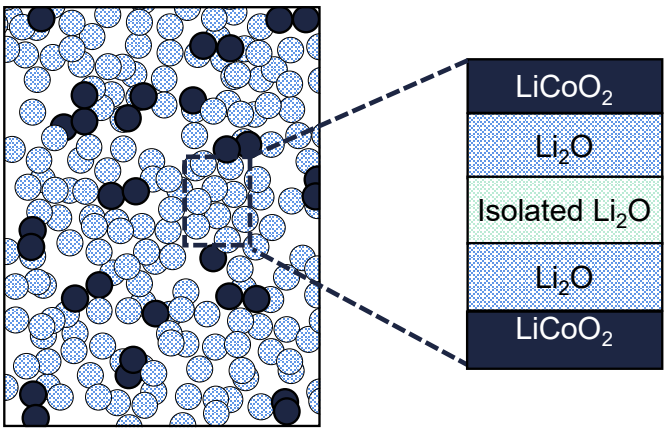

Figure. 7 Schematic diagram of the origin of low extracted Li ratio when $x(Li_2O) > 0.7$.

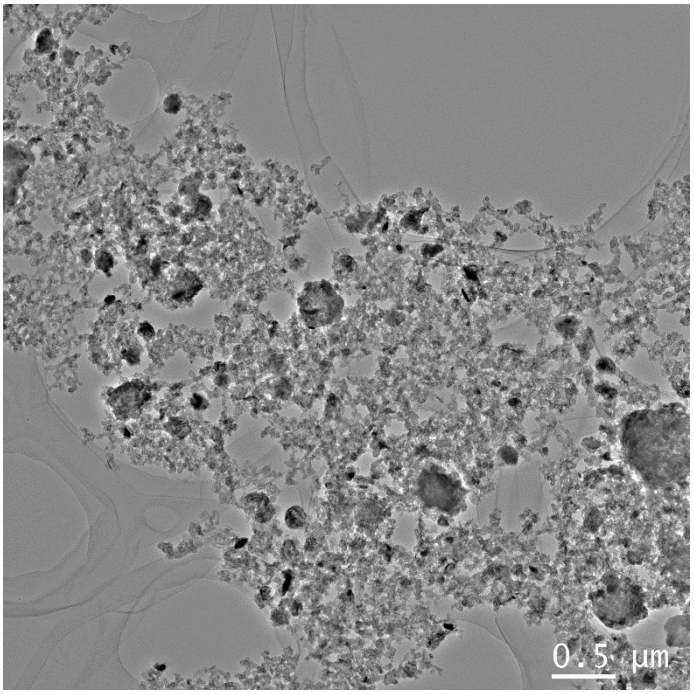

Figure. 8 TEM image of charged Co-Li_2O. The dark dots are remained $Li_{1-x}CoO_2$ species, the particles around dark dots are conductive carbon black.

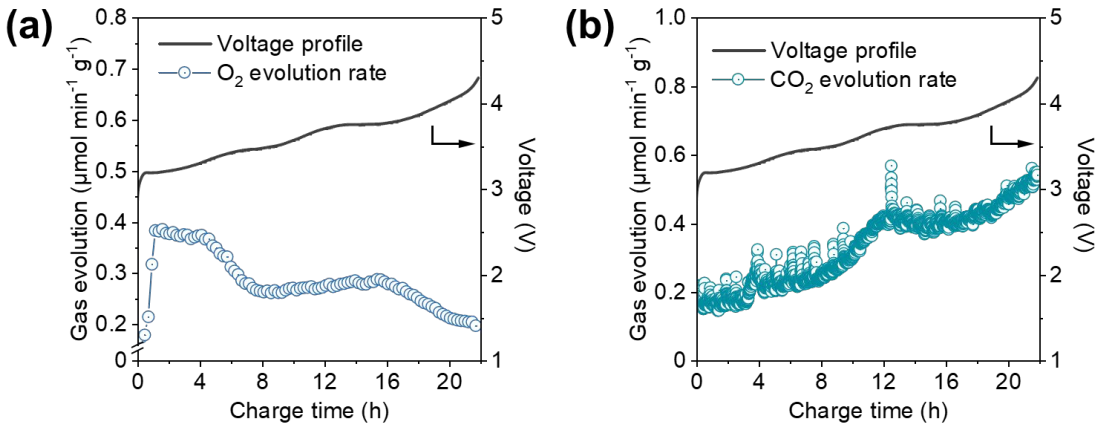

Figure. 9 Gas evolution rate and correspondent voltage profile of the Co-Li_2O electrode during charging. (a) O_2 and (b) CO_2.

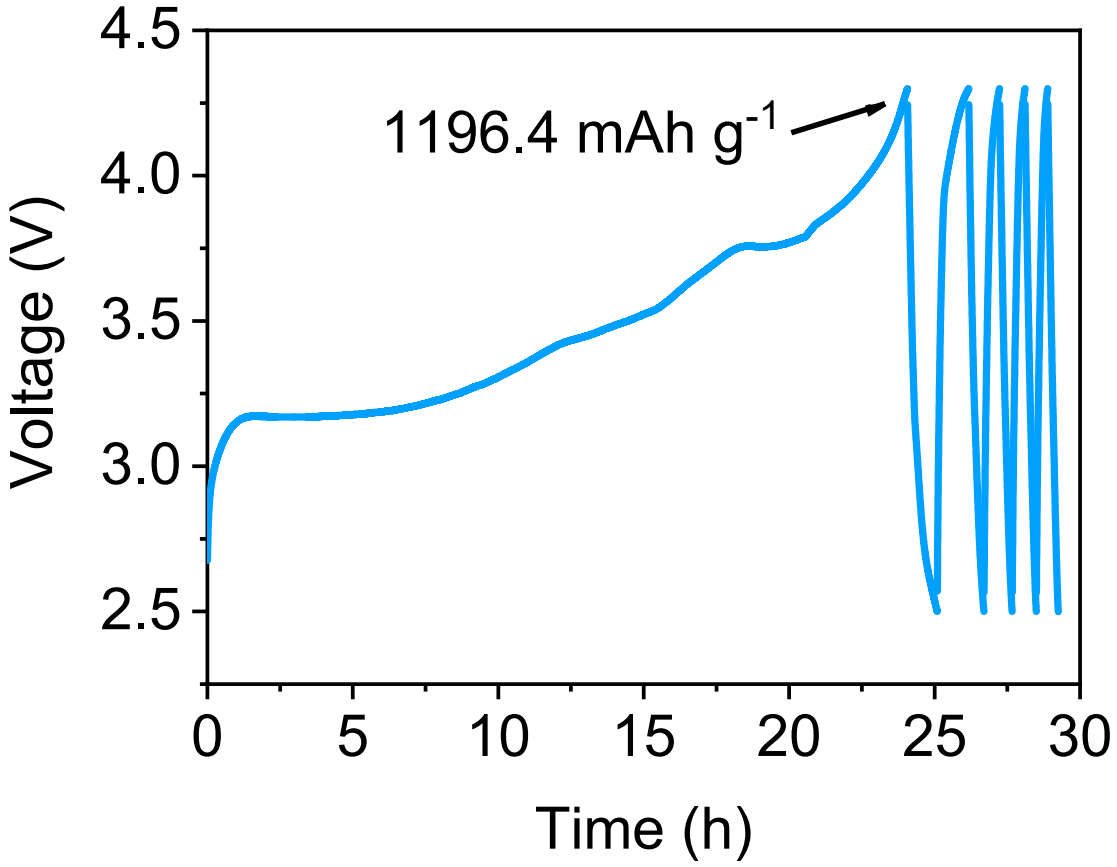

Figure. 10 Initial galvanostatic charge-discharge profiles of Co-Li_2O using Li_2O and wasted $LiCoO_2$ as raw materials.

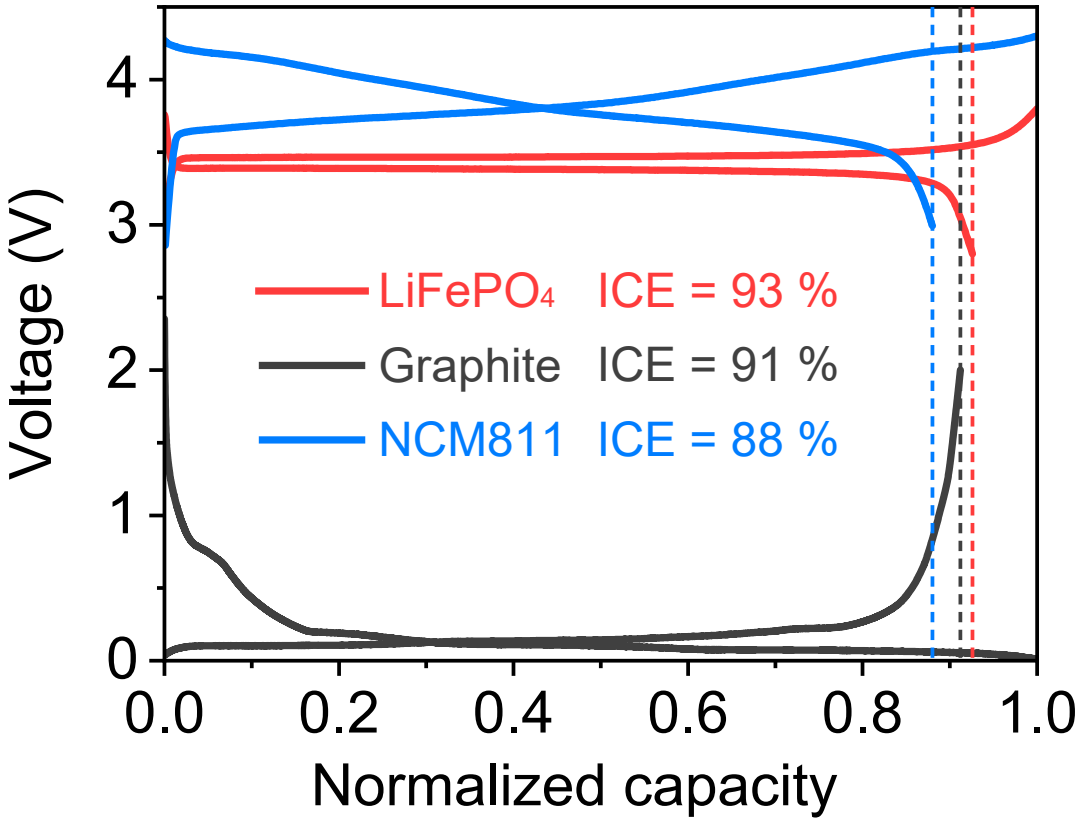

Figure. 11 ICE comparison of $LiFePO_4$, graphite and NCM811.

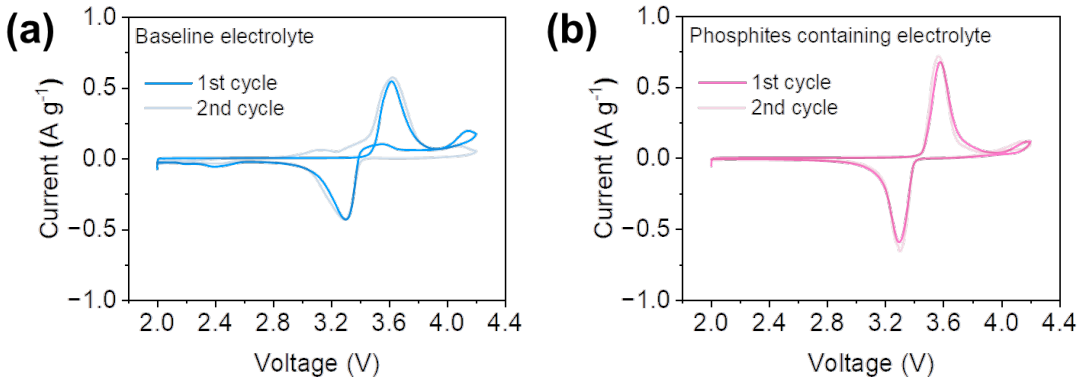

Figure. 12 (a) CV curves of $LiFePO_4$ electrode with 3.5% Co-Li_2O in baseline electrolyte. (b) CV curves of $LiFePO_4$ electrode with 3.5% Co-Li_2O in phosphites-containing electrolyte. The scanning rate is 0.2 $mV \cdot s^{-1}$.

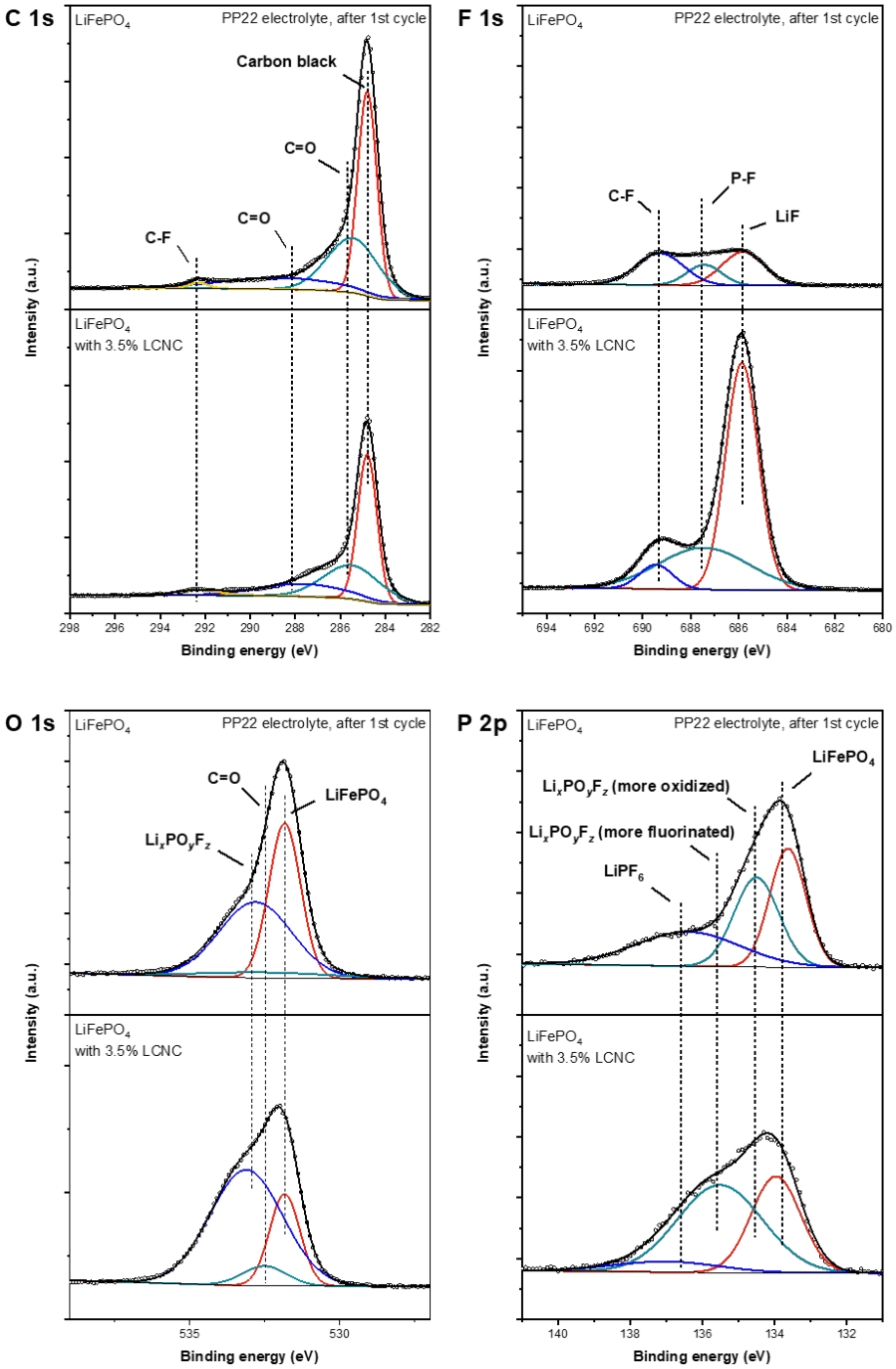

Figure .13 XPS of charged $LiFePO_4$ electrode with 3.5% Co-Li_2O and without additive in phosphites-containing electrolyte. Here the phosphites-containing electrolyte was abbreviated to PP22 for convenience.

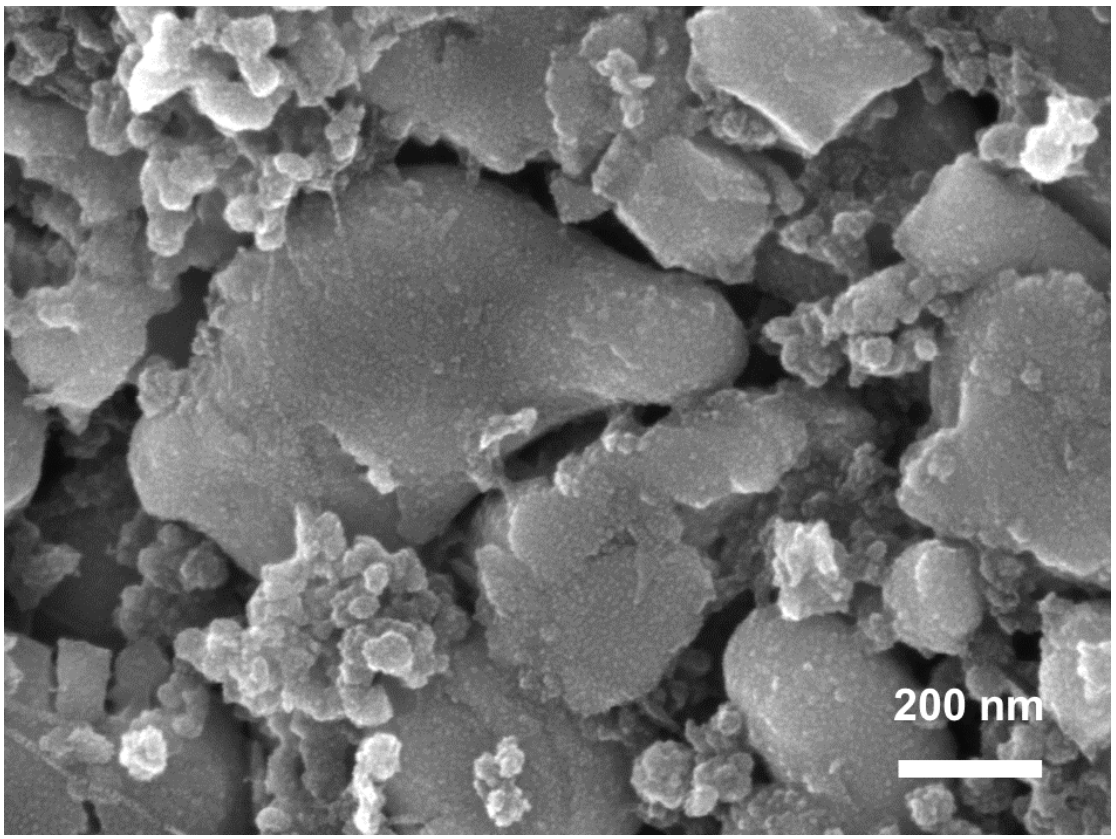

Figure. 14 Cross-sectional SEM image of charged $LiFePO_4$ electrode with 3.5% Co-Li_2O.

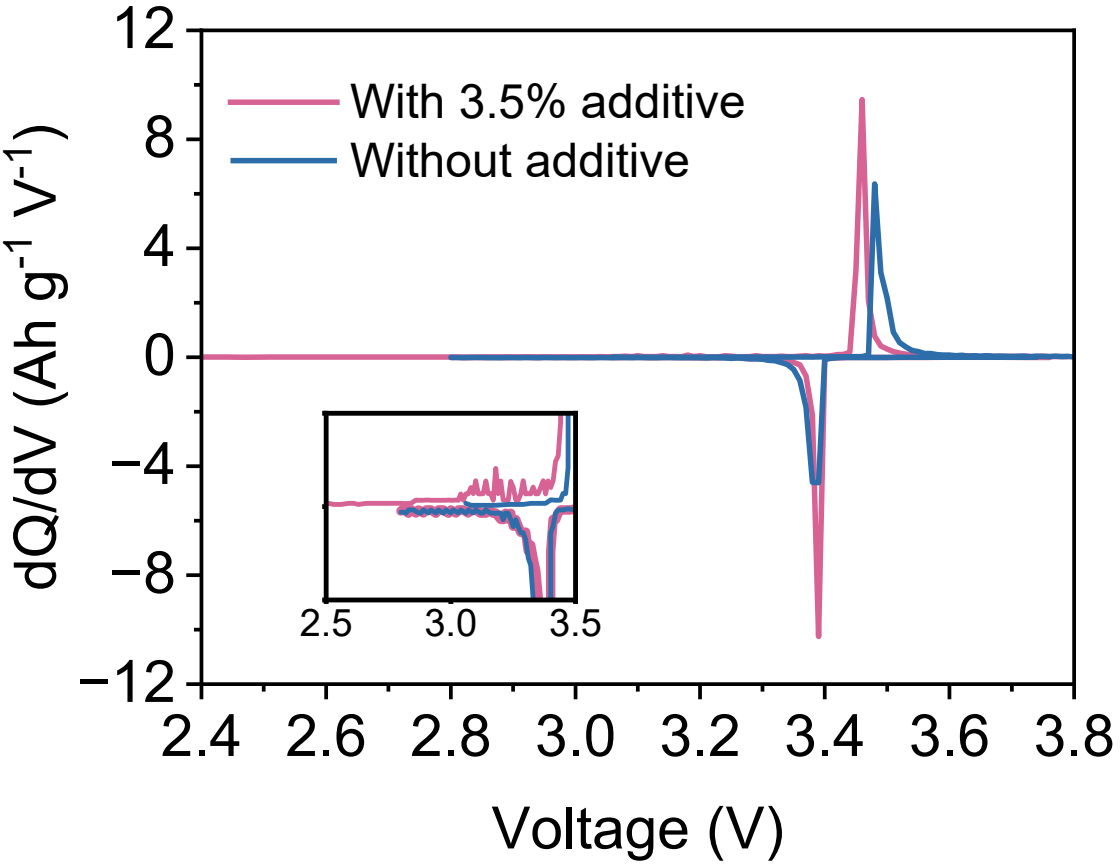

Figure. 15 First cycle dQ/dV plots of Li || $LiFePO_4$ full cells with and without 3.5% Co-Li_2O.

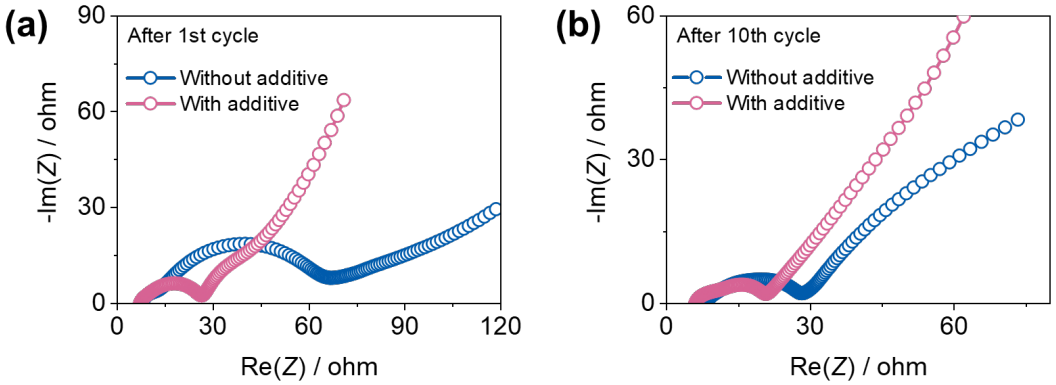

Figure. 16 Electrochemical impedance spectra with and without additive. (a) After the first cycle. (b) After 10 cycle.

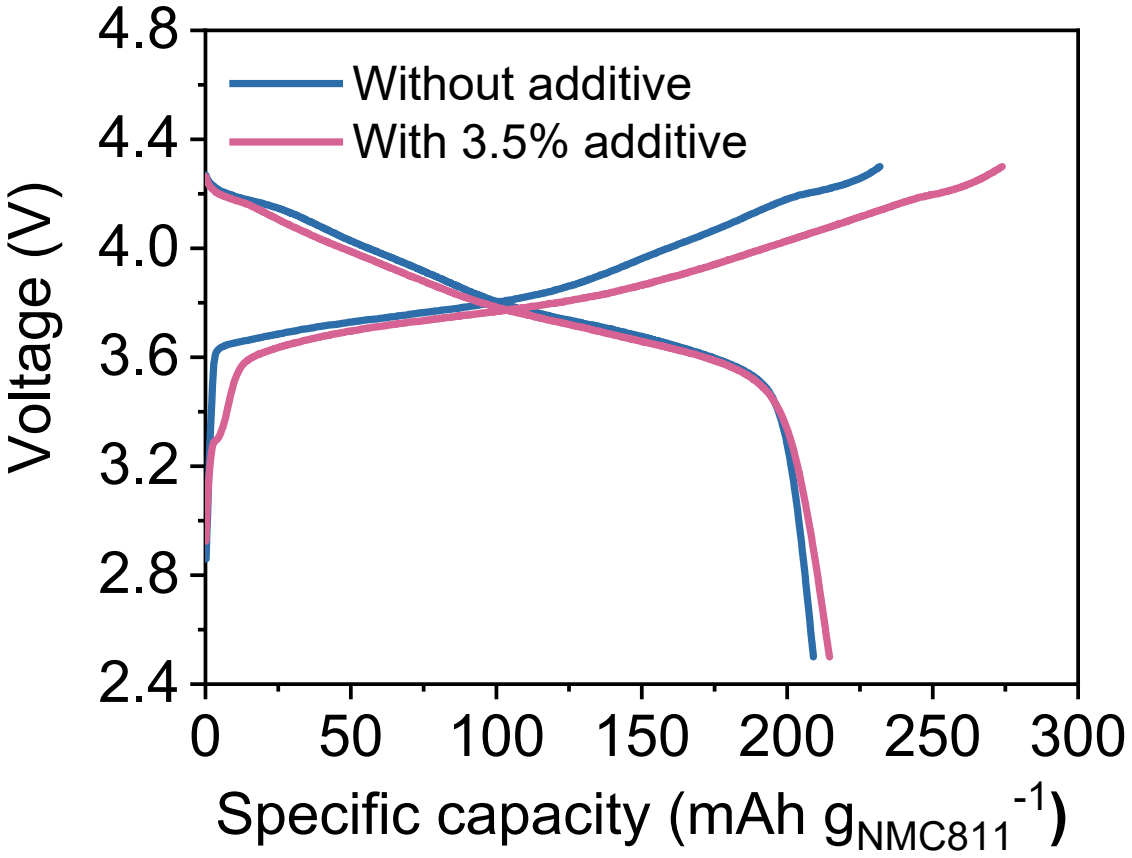

Figure. 17 Voltage profiles of $LiNi_{0.8}Co_{0.1}Mn_{0.1}O_2$ half cells with and without 3.5% Co-Li_2O. The current density is 50 mA· g_{NMC811}^{-1}.

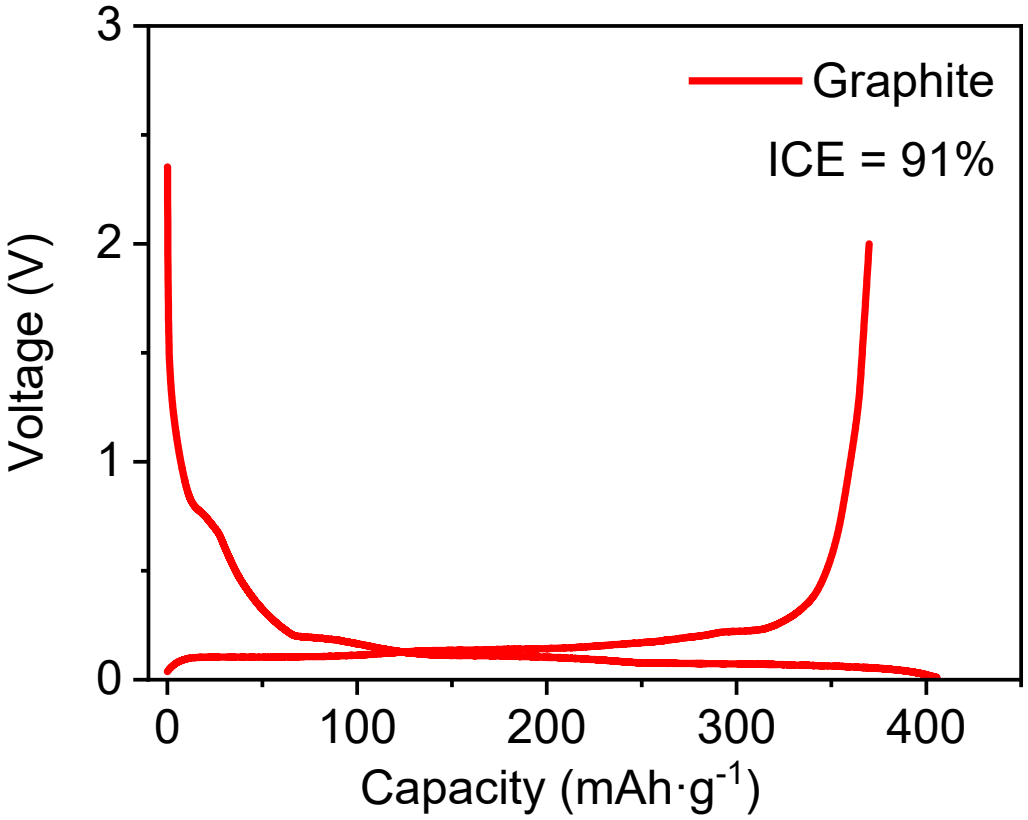

Figure. 18 Initial charge-discharge profile of graphite.

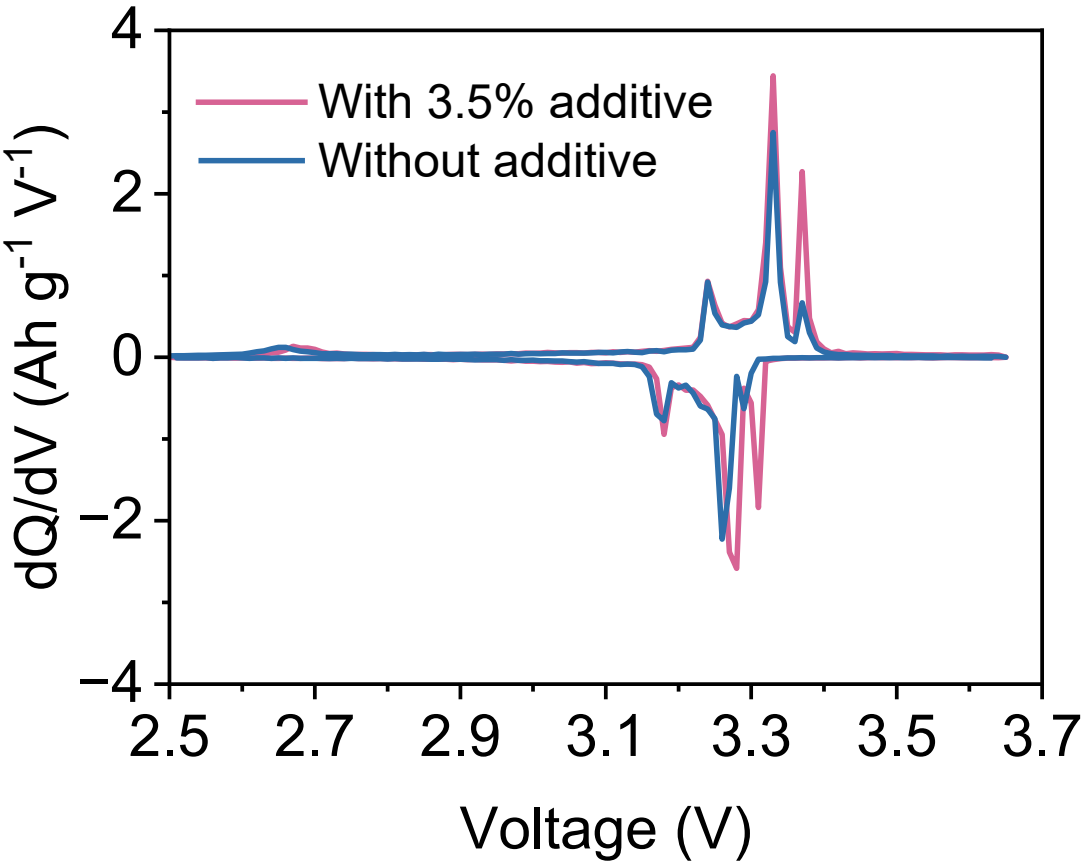

Figure. 19 First cycle dQ/dV plots of graphite || $LiFePO_4$ full cells with and without 3.5% Co-Li_2O.