Supplementary Materials

Ni-Rich Layered Oxide with Preferred Orientation (110) Plane as a Stable Cathode Material for High-Energy Lithium-Ion Batteries

Fangkun Li, Zhengbo Liu, Jiadong Shen, Xijun Xu, Liyan Zeng, Yu Li, Dechao Zhang, Shiyong Zuo and Jun Liu*

Guangdong Provincial Key Laboratory of Advanced Energy Storage Materials, School of Materials Science and Engineering, South China University of Technology, Guangzhou 510641, China; mslifk@scut.edu.cn (F.L.); 201810103813@mail.scut.edu.cn (Z.L.); 201910103734@mail.scut.edu.cn (J.S.); xuxijun2019@scut.edu.cn (X.X.); 201820117753@mail.scut.edu.cn (L.Z.); 201820117766@mail.scut.edu.cn (Y.L.); 201810103808@mail.scut.edu.cn (D.Z.); shyzuo@scut.edu.cn (S.Z.)

* Correspondence: msjliu@scut.edu.cn

Figure S1. SEM image and elemental EDS mapping of Ni, Co, Mn, O and all elements for the intermediate oxides composite, the scale bar is 90 μm in all figures.
Figure S2. EDS spectrum and corresponding element composition of intermediate oxides composite.

| Element | Wt%  | At%  |
|---------|------|------|
| O       | 20.61| 23.32|
| Ni      | 22.30| 6.88 |
| Co      | 7.21 | 2.22 |
| Mn      | 6.47 | 2.13 |

Figure S3. Applied current plus vs. cell voltage for a single titration step of GITT curves.
Figure S4. N\textsubscript{2} adsorption/desorption isotherms of NCM622 nanobricks.

Figure S5. Typical XRD pattern and SEM images of NCM622 electrode after long-term 200 cycles at 0.5 C rate.

Figure S6. Equivalent circuit model is used for fitting the experimental results. $R_s$: solution resistance, $R_f$: surface film resistance, related to Li-ions diffusion in the cathode electrolyte interface (CEI), and $R_{ct}$: charge transfer resistance, $CPE$: constant phase element, $W_o$: Warburg element (open).
Table S1. Unit cell parameters for the two fundamental phases of Ni(OH)$_2$.

|                  | α-Ni(OH)$_2$ | β-Ni(OH)$_2$ |
|------------------|--------------|--------------|
| Space group      | $D_{3d}^{1}/P_{3}^3Im/No.162$ | $D_{3d}^{1}/P_{3}^3Im/No.164$ |
| $a = b$          | 3.08 Å       | 3.126 Å      |
| $c$              | 8.0 Å        | 4.605 Å      |
| $\alpha = \beta = 90^\circ$, $\gamma = 120^\circ$ | |

Table S2. X-ray diffraction parameters of α-Ni(OH)$_2$ based on JCPDS No.38-0715.

| Miller indices (hkl) | d (Å) | 2θ (°) | l (a. u.) |
|----------------------|-------|--------|-----------|
| (003)                | 7.79  | 11.349 | 100.0     |
| (006)                | 3.91  | 22.735 | 70.0      |
| (101)                | 2.68  | 33.458 | 50.0      |
| (012)                | 2.60  | 34.412 | 50.0      |
| (015)                | 2.32  | 38.77  | 50.0      |
| (018)                | 1.97  | 45.99  | 20.0      |
| (110)                | 1.54  | 59.98  | 50.0      |
| (113)                | 1.51  | 61.25  | 20.0      |

Table S3. X-ray diffraction parameters of β-Ni(OH)$_2$ based on JCPDS No. 14-0117.

| Miller indices (hkl) | d (Å) | 2θ (°) | l (a. u.) |
|----------------------|-------|--------|-----------|
| (001)                | 4.61  | 19.258 | 100.0     |
| (100)                | 2.71  | 33.064 | 45.0      |
| (101)                | 2.33  | 38.541 | 100.0     |
| (002)                | 2.30  | 39.098 | 2.2       |
| (012)                | 1.75  | 52.100 | 35.0      |
| (110)                | 1.56  | 59.052 | 25.0      |
| (003)                | 1.53  | 60.240 | <1        |
| (111)                | 1.48  | 62.73  | 16.0      |

Table S4. The ICP-OES results of NCM622 nanobricks.

| Li   | Ni   | Co   | Mn   |
|------|------|------|------|
| Weight ratio (%) | 6.97 | 35.03 | 11.50 | 11.12 |
| Molar ratio (%)   | 1.03 | 0.61 | 0.20 | 0.20 |

Table S5. Atomic site information and crystallographic data for NCM622.

| Atom | Wyck. | a | b   | c   | Occ.  | $U_i/U_e \times 100$ |
|------|-------|---|-----|-----|-------|---------------------|
| Li1  | 3a    | 0 | 0   | 0.5 | 0.946 | 1.89                |
| Ni2  | 3a    | 0 | 0   | 0   | 0.054 | 1.89                |
| Li2  | 3b    | 0 | 0   | 0.5 | 0.010 | 0.70                |
| Ni1  | 3b    | 0 | 0   | 0.5 | 0.590 | 0.70                |
| Co1  | 3b    | 0 | 0   | 0.5 | 0.200 | 0.70                |
| Mn1  | 3b    | 0 | 0   | 0.5 | 0.200 | 0.70                |
| O2   | 6c    | 0 | 0   | 0.2566(12) | 1.000 | 0.54                |

Lattice parameters (Å) $a = b = 2.86770(4)$, $c = 14.21426(27)$, $\alpha = \beta = 90^\circ$, $\gamma = 120^\circ$

Cell volume 101.2333(28) Å$^3$