Supporting Information

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Morphological and Structural Characterization of Mg-substituted Ni-rich NMC Materials

Figure S1. High-magnification SEM micrographs of lithiated products to reveal changes in shape and size of primary particles: a) NMC900505, b) NMCMg 90040501, c) NMCMg 9003.50501.5, d) NMCMg 90030502, and e) NMCMg 90050401.

Table S1. Particle size distribution (by volume) and tap density of the co-precipitated hydroxide precursors.

| Targeted stoichiometry | Material       | Tap density (g cm$^{-3}$) | $d_{50}$ (µm) | $d_{90}$ (µm) | $d_{mean}$ (µm) |
|------------------------|----------------|----------------------------|----------------|---------------|------------------|
| Ni$_{0.90}$Mn$_{0.05}$Co$_{0.05}$Mg$_{0.01}$ (OH)$_2$ | NMCMg 90050401 | 1.81 ± 0.10                | 12.6 ± 0.3     | 22.4 ± 0.2     | 13.4 ± 0.3       |
| Ni$_{0.90}$Mn$_{0.05}$Co$_{0.05}$Mg$_{0.01}$ (OH)$_2$ | NMCMg 90030502 | 1.85 ± 0.20                | 12.3 ± 0.4     | 20.7 ± 0.4     | 12.9 ± 0.4       |
| Ni$_{0.90}$Mn$_{0.05}$Co$_{0.05}$Mg$_{0.01}$ (OH)$_2$ | NMCMg 900350501 | 1.83 ± 0.15                | 11.9 ± 0.2     | 22.4 ± 0.1     | 12.8 ± 0.1       |
| Ni$_{0.90}$Mn$_{0.05}$Co$_{0.05}$Mg$_{0.01}$ (OH)$_2$ | NMCMg 900040501 | 1.80 ± 0.20                | 11.1 ± 1.1     | 20.7 ± 2.7     | 11.9 ± 1.4       |
| Ni$_{0.90}$Mn$_{0.05}$Co$_{0.05}$Mg$_{0.01}$ (OH)$_2$ | NMCMg 9000505 | 1.83 ± 0.10                | 12.2 ± 0.8     | 21.0 ± 1.3     | 12.6 ± 0.7       |
| Ni$_{0.90}$Mn$_{0.05}$Co$_{0.05}$Mg$_{0.01}$ (OH)$_2$ | NMC 801010 | 1.79 ± 0.10                | 10.8 ± 0.6     | 18.2 ± 1.2     | 10.8 ± 0.7       |
| Ni$_{0.80}$Mn$_{0.10}$Co$_{0.10}$ (OH)$_2$ | NMC 801010 | 1.79 ± 0.10                | 12.2 ± 0.8     | 21.0 ± 1.3     | 12.6 ± 0.7       |
| Ni$_{0.90}$Mn$_{0.05}$Co$_{0.05}$Mg$_{0.01}$ (OH)$_2$ | NMCMg 900350501 | 1.83 ± 0.15                | 11.9 ± 0.2     | 22.4 ± 0.1     | 12.8 ± 0.1       |
| Ni$_{0.90}$Mn$_{0.05}$Co$_{0.05}$Mg$_{0.01}$ (OH)$_2$ | NMCMg 90030502 | 1.85 ± 0.20                | 12.3 ± 0.4     | 20.7 ± 0.4     | 12.9 ± 0.4       |
| Ni$_{0.90}$Mn$_{0.05}$Co$_{0.05}$Mg$_{0.01}$ (OH)$_2$ | NMCMg 90050401 | 1.81 ± 0.10                | 12.6 ± 0.3     | 22.4 ± 0.2     | 13.4 ± 0.3       |
Figure S2. SEM micrographs of NMC 801010 a) hydroxide-based precursor (\(TM(\text{OH})_2\)) and b) lithiated product (\(\text{LiTMO}_2\); \(TM=\text{Ni, Co and Mn}\)). High-magnification SEM micrographs shown in the inset of the figures reveal shape and size of primary particles. c) Particle size distribution (by volume) of co-precipitated hydroxide precursors and lithiated products.
Table S2. Rietveld refinement results for XRD experiments on lithiated Mg-substituted Ni-rich cathode powdered materials: R-weighted pattern (R$_{wp}$), R-expected (R$_{exp}$). Goodness of fit (GOF), lattice parameters $a$ and $c$, unit cell volume (V), Li/Ni mixing, z-Position and isotropic thermal factors. Six different hypotheses for placement of Mg$^{2+}$ ions inside the NMC structure were considered: i) w/o considering Mg; ii) Mg only occupying 3$b$ sites (transition metals (TM$s$) layer); iii) Mg occupying both 3$a$ (Li layer) and 3$b$ sites (TM$s$ layer); iv) Mg occupying 3$a$ sites (Li layer) and Li on 3$b$ sites (TM$s$ layer); v) Mg occupying 3$a$ sites (Li layer) with vacancies on 3$b$ sites (TM layer) and vi) Mg occupying 3$a$ sites (Li layer) with TM$=1$. Rietveld refinement results shown here for each sample were chosen for the hypothesis better fitting the experimental results based on the R$_{wp}$ and the GOF of refinements, i.e., lower GOF and R$_{wp}$.

| Material      | NMC 801010 | NMC 900505 | NMCMg 90040501 | NMCMg 9003.50501.5 | NMCMg 90030502 | NMCMg 90050401 |
|---------------|------------|------------|----------------|-------------------|----------------|----------------|
| Hypothesis for the refinement | w/o Mg | w/o Mg | Mg on TM and Li sites | Mg on Li, Li, Li on TM | Mg on Li on TM | Mg on Li, Li on TM |
| R$_{wp}$      | 1.37       | 1.85       | 1.95           | 1.62              | 1.30           | 1.81           |
| R$_{exp}$     | 0.85       | 0.98       | 1.03           | 0.96              | 0.72           | 0.97           |
| GOF           | 1.61       | 1.89       | 1.89           | 1.69              | 1.81           | 1.87           |
| $a / 10^3$Åm | 2.8717(5)  | 2.8758(3)  | 2.8755(3)     | 2.8747(2)         | 2.8749(2)     | 2.8766(4)     |
| $c / 10^3$Åm | 14.208(4)  | 14.204(3)  | 14.203(3)     | 14.202(2)         | 14.301(2)     | 14.206(3)     |
| V / 10$^3$Åm$^3$ | 101.47(5) | 101.73(3)  | 101.71(3)    | 101.64(2)         | 101.65(2)    | 101.80(3)    |
| $c/a$         | 4.9480(2)  | 4.9389(5)  | 4.9392(5)     | 4.9402(4)         | 4.9397(4)    | 4.9385(5)    |
| Li / Ni mixing / % | 3.1 ± 0.1  | 4.1 ± 0.1  | 3.6 ± 0.1     | 3.0 ± 0.1         | 2.6 ± 0.1    | 4.6 ± 0.1    |
| z (O)         | 0.24(1)    | 0.24(1)    | 0.24(1)       | 0.24(1)           | 0.24(1)      | 0.24(1)      |
| beq(O)        | 0.7(5)     | 0.9(4)     | 0.6(5)        | 0.5(4)            | 0.6(3)       | 0.9(4)       |
| beq(TM)       | 0.8(2)     | 0.4(2)     | 0.3(2)        | 0.3(2)            | 0.3(1)       | 0.6(2)       |
Figure S3. Detailed Rietveld refinements of XRD patterns of Ni-rich Mg-substituted NMC cathode powdered materials: a) NMC 801010, b) NMC 900505, c) NMCMg 90040501, d) NMCMg 9003.50501.5, e) NMCMg 90030502 and f) NMCMg 90050401. The panels on the right show expanded views of the (006)/(102) and (018)/(110) reflections slit up as an indication of well-defined layered structure. The black empty dots show the original data points, the solid red lines the calculated results while the blue lines show the differences between the measured and calculated data. R-weighted pattern (R_{wp}), R-expected (R_{exp}) and GOF are shown in the inset. Rietveld refinement results shown here for each sample were chosen for the hypothesis for placement of Mg^{2+} ions inside the NMC structure better fitting the experimental results based on the R_{wp} and the GOF of refinements, i.e., lower GOF and R_{wp}. 
Figure S4. Detailed results of a) goodness of fit (GOF) of Rietveld refinements based on the different hypotheses for placement of Mg$^{2+}$ ions inside the NMC structure: i) w/o considering Mg; ii) Mg only occupying 3$b$ sites (transition metals (TMs) layer); iii) Mg occupying both 3$a$ (Li layer) and 3$b$ sites (TMs layer); iv) Mg occupying 3$a$ sites (Li layer) and Li on 3$b$ sites (TMs layer); v) Mg occupying 3$a$ sites (Li layer) with vacancies on 3$b$ sites (TM layer) and vi) Mg occupying 3$a$ sites (Li layer) with TM=1. b) Evolution of Li$^+$/Ni$^{2+}$ mixing for Ni-rich Mg-substituted NMC cathode powdered materials.
**Electrochemical Properties of NMC || Li Metal Cells and NMC || Graphite Cells**

![Graph of C-rate performance](image)

**Figure S5.** C-rate performance of Mg-substituted Ni-rich cathode materials in NMC || Li metal cells: a) specific discharge capacity vs. cycle number and b) capacity retention with respect to the 1st discharge capacity at 0.1C for each material. Cell voltage range: 2.9 - 4.3, 2.9 - 4.4 and 2.9 - 4.5 V. Error bars: standard deviation of three cells for each sample. Electrolyte: 1 M LiPF$_6$ in 3:7 vol. % EC/EMC + 2 wt.% VC.
Figure S6. 1st cycle a) cell voltage profiles and b) corresponding differential capacity plots (dQ/dV) vs. voltage plots of NMC || Li-metal cells at 20 °C within a cell voltage range of 2.9–4.3 V at 0.1C. Electrolyte: 1 M LiPF₆ in 3:7 vol. % EC/EMC + 2 wt.% VC. H and M refer to hexagonal and monoclinic phases and the different phase transitions are represented as H₁-M, the M-H₂, and H₂-H₃ in the corresponding transition peaks.
Figure S7. 1st cycle Coulombic efficiencies ($C_{Efi}$) of a) NMC || Li-metal cells and b) NMC || graphite full-cells as a function of Mg content at 0.1C. Error bars represent the standard deviations of three cells tested for each sample. Cell voltage range for NMC || Li: 2.9 - 4.3 V. Cell voltage range for NMC || graphite: 2.8 - 4.2 V. Electrolyte: 1 M LiPF$_6$ in 3:7 vol. % EC/EMC + 2 wt.% VC.
| Cathode stoichiometry | Electrode composition (Active material:binder: conductive agent) | Experimental conditions | Specific discharge capacity / mAh g\(^{-1}\) | Cycle life | Reference |
|----------------------|---------------------------------------------------------------|-------------------------|-----------------------------------------------|------------|-----------|
| LiNi\(_{0.90}\)Co\(_{0.04}\)Mn\(_{0.05}\)Mg\(_{0.01}\)O\(_2\) (NMCMg 90040501) | 94:03:03 | Set-up: NMCMg||graphite two-electrode set-up Electrolyte: 1 M LiPF\(_6\) in 3:7 vol.% EC/EMC + 2 wt.% VC Cathode active mass loading: 12.0±0.5 mg cm\(^{-2}\) Cell voltage window: 2.8 - 4.2 V N/P balancing ratio: 1.15:1.00 | ≈ 191.8 ± 0.1 mAh g\(^{-1}\) at 0.1C (1C = 190 mA g\(^{-1}\)) | 361 cycles to 80% state-of-health (SOH) (long-term cycling at 0.33C) | This work |
| LiNi\(_{0.90}\)Co\(_{0.02}\)Mn\(_{0.05}\)Mg\(_{0.02}\)O\(_2\) (NMCMg 90030502) | 94:03:03 | Set-up: NMCMg||graphite two-electrode set-up Electrolyte: 1 M LiPF\(_6\) in 3:7 vol.% EC/EMC + 2 wt.% VC Cathode active mass loading: 12.0±0.5 mg cm\(^{-2}\) Cell voltage window: 2.8 - 4.2 V N/P balancing ratio: 1.15:1.00 | ≈ 183 ± 1 mAh g\(^{-1}\) at 0.1C (1C = 190 mA g\(^{-1}\)) | 607 cycles to 80% SOH (long-term cycling at 0.33C) | This work |
| Li\(_{0.98}\)Ni\(_{0.90}\)Co\(_{0.06}\)Mg\(_{0.02}\)O\(_2\) | 80:10:10 | Set-up: NCMg||graphite two-electrode set-up Electrolyte: 1 M LiPF\(_6\) in 3:7 vol.% EC/EMC + 2 wt.% VC Cathode active mass loading: 5 mg cm\(^{-2}\) Cell voltage window: 2.5 - 4.3 V N/P balancing ratio: 1.08:1.00 | ≈ 207 mAh g\(^{-1}\) at 0.1C (1C = 180 mA g\(^{-1}\)) | 500 cycles to 80% SOH (long-term cycling at 0.5C) | [1] |
| Li(Ni \(_{0.8}\)Co\(_{0.1}\)Mn\(_{0.1}\))\(_{0.97}\)Mg\(_{0.03}\)O\(_2\) | 80:10:10 | Set-up: NCMg||Li two-electrode set-up Electrolyte: 1.15 M 1 LiPF\(_6\) in 1:2:2 vol.% EC:EMC:DMC Cathode active mass loading: 3.0 mg cm\(^{-2}\) Cell voltage window: 3.0 - 4.5 V N/P balancing ratio: ---- | ≈ 226.5 mAh g\(^{-1}\) at 0.1C (1C = 190 mA g\(^{-1}\)) | 350 cycles to 80% SOH (long-term cycling at 0.5C) | [2] |
| Cathode stoichiometry (Active material:binder: conductive agent) | Electrode composition | Experimental conditions | Specific discharge capacity / mAh g⁻¹ | Cycle life | Reference |
|---------------------------------------------------------------|-----------------------|-------------------------|----------------------------------------|------------|-----------|
| LiNi₀.₉₅Mg₀.₀₅O₂                                              | 92:04:04              | Set-up: NMg||Li two-electrode set-up | ≈ 200 mAh g⁻¹ at 0.05C (1C = 200 mA g⁻¹) | 100 cycles to ≈96% SOH (long-term cycling at 0.2C) | [3] |
| LiNi₀.₉₀Co₀.₀₇Mg₀.₀₃O₂                                          | 85:7.5:7.5            | Set-up: NCMg||Li two-electrode set-up | ≈ 224.4 mAh g⁻¹ at 0.1C (1C = 180 mA g⁻¹) | 300 cycles to 84% SOH (long-term cycling at 2C) | [4] |
| LiNi₀.₉₆Mn₀.₀₂Mg₀.₀₂O₂                                          | 90:05:05              | Set-up: NMMg||Li two-electrode set-up | ≈ 220 mAh g⁻¹ at 0.1C (1C = 200 mA g⁻¹) | 350 cycles to 80% SOH (long-term cycling at 0.33C) | [5] |
Figure S8. Evolution of a) specific energies at material level based on cathode active mass and b) accumulated Coulombic inefficiencies (ACI) vs. cycle number of NMC ∥ graphite full-cells. Error bars represent the standard deviation of three cells tested for each sample. Cell voltage range: 2.8 - 4.2 V. Electrolyte: 1 M LiPF$_6$ in 3:7 vol. % EC/EMC + 2 wt.% VC. The specific energy at material level ($E_g$) was calculated from the mean discharge voltages ($U_{dis}$), discharge capacities ($Q_{dis}$) and cathode active mass according to the equation: $E_g = \frac{U_{dis} Q_{dis}}{m_{cathode}}$. 
Table S4. Initial specific capacity at 0.1C and 0.33C, mean discharge voltage, calculated initial specific energy, and cycle life to 80 % SOH of NMC || graphite full-cells. Cell voltage range: 2.8 - 4.2 V. Electrolyte: 1 M LiPF$_6$ in 3:7 vol. % EC/EMC + 2 wt.% VC.

| Material       | NMC 801010 | NMC 900505 | NMC 90040501 | NMC 900350501.5 | NMC 90030502 | NMC 90050401 |
|----------------|------------|------------|---------------|-----------------|--------------|--------------|
| Initial discharge capacity at 0.1C (2$^{nd}$ cycle) / mAh g$^{-1}$ | 185.4 ± 0.4 | 200.3 ± 0.4 | 191.8 ± 0.10 | 190.4 ± 0.1 | 183.2 ± 1.4 | 184.2 ± 0.1 |
| Initial discharge capacity at 0.33C (5$^{th}$ cycle) / mAh g$^{-1}$ | 177.8 ± 0.2 | 193.6 ± 0.3 | 184.9 ± 0.1 | 183.4 ± 0.4 | 175.4 ± 1.2 | 176.9 ± 0.3 |
| Mean discharge voltage at 0.1C (2$^{nd}$ cycle) / V | 3.712 ± 0.001 | 3.71 ± 0.02 | 3.72 ± 0.01 | 3.73 ± 0.01 | 3.732 ± 0.004 | 3.730 ± 0.004 |
| Mean discharge voltage at 0.33C (5$^{th}$ cycle) / V | 3.714 ± 0.001 | 3.69 ± 0.02 | 3.71 ± 0.01 | 3.71 ± 0.03 | 3.718 ± 0.007 | 3.715 ± 0.001 |
| Specific energy at 0.1C (2$^{nd}$ cycle) / Wh kg$^{-1}$ | 688.4 ± 0.3 | 745.6 ± 2.4 | 714.1 ± 0.6 | 709.8 ± 0.5 | 683.9 ± 5.2 | 687.0 ± 0.1 |
| Specific energy at 0.33C (5$^{th}$ cycle) / Wh kg$^{-1}$ | 660.8 ± 0.9 | 717.3 ± 2.7 | 686.0 ± 1.2 | 681.5 ± 2.1 | 652.3 ± 5.6 | 657.4 ± 0.8 |
| Cycle life to 80% SOH | ≈512 | ≈229 | ≈361 | ≈411 | ≈607 | ≈329 |
Figure S9. Accumulated discharge specific energies for NMC 900505, NMCMg 90040501 and NMCMg 90030502 samples for a) first 50 cycles and b) 250 cycles upon long-term cycling in NMC||graphite full-cells. Only representative samples are shown to better distinguish differences between samples. Red arrows point out the differences in energy throughput through the material in the first cycles and the cycle where the NMCMg 90030502 sample outperforms the reference NMC 900505 sample, respectively.
Post-mortem ex-situ XRD investigations

Table S5. Pawley refinement results for XRD experiments on pristine, charged (up to 4.2 V or to 200 mAh g⁻¹ corresponding to x=0.72 in Li₁₋ₓTMO₂) and discharged (2.8 V – after charging to 4.2 V) Ni-rich cathode electrodes: R-weighted pattern (R_wp), R-expected (R_exp), Goodness of fit (GOF), lattice parameters a and c, and unit cell volume (V). Average values along with standard deviations from two refinements are listed for the pristine and charged electrodes. The XRD pattern of the sample NMC 900505 was fitted to two different phases to account for the two-phase reflections observed in the charge state. However, for the other samples only one phase was fitted, as there was no clear reflection splitting.

| Material               | NMC 801010 | NMC 900505 | NMCMg 90040501 | NMCMg 9003.50501.5 | NMCMg 90030502 | NMCMg 90050401 |
|------------------------|------------|------------|-----------------|-------------------|----------------|----------------|
| **Pristine electrodes**|            |            |                 |                   |                |                |
| R_wp                   | 1.14       | 1.33       | 1.55            | 1.44              | 1.68           | 1.26           |
| R_exp                  | 0.80       | 0.84       | 0.82            | 0.83              | 0.82           | 0.85           |
| GOF                    | 1.42       | 1.58       | 1.89            | 1.72              | 2.07           | 1.48           |
| \(a/10\text{Å}m\)      | 2.871(1)   | 2.87512(3) | 2.8747(5)       | 2.8739(5)         | 2.8741(2)      | 2.8758(2)      |
| \(c/10\text{Å}m\)      | 14.20(1)   | 14.192(1)  | 14.19(2)        | 14.188(1)         | 14.19(2)       | 14.196(6)      |
| \(V/10^{30}\text{m}^3\)| 101.4(2)   | 101.6(1)   | 101.5(2)        | 101.49(5)         | 101.50(2)      | 101.67(4)      |
| \(c/a\)               | 4.9451(4)  | 4.9362(5)  | 4.9355(5)       | 4.9369(4)         | 4.9366(4)      | 4.9362(5)      |
| **Charged electrodes** |            |            |                 |                   |                |                |
| R_wp                   | 2.22       | 2.01       | 3.69            | 3.39              | 3.65           | 3.22           |
| R_exp                  | 0.805      | 0.83       | 0.815           | 0.83              | 0.81           | 0.855          |
| GOF                    | 2.77       | 2.43       | 4.525           | 4.08              | 4.52           | 3.775          |
| \(a/10\text{Å}m\)      | 2.818(1)   | 2.820(5)   | 2.8214(7)       | 2.820(4)          | 2.8228(4)      | 2.8262(9)      |
| \(c/10\text{Å}m\)      | -----      | 2.82(2)    | -----           | -----             | -----          | -----          |
| \(V/10^{30}\text{m}^3\)| ----       | 14.02(2)   | 14.3(5)         | 14.2(1)           | 14.27(5)       | 14.3(5)        |
| \(c/a\)               | 5.0863(3)  | 4.970(2)   | 5.0577(5)       | 5.0522(3)         | 5.0544(3)      | 5.0457(5)      |
### Charged electrodes
200 mAh g$^{-1}$ (x=0.72 in Li$_{1-x}$TMO$_2$)

| Material        | NMC 801010 | NMC 900505 | NMC 90040501 | NMC 9003.50501.5 | NMC 90030502 | NMC 90050401 |
|-----------------|------------|------------|--------------|------------------|--------------|--------------|
| $R_{wp}$        | -----      | 2.01       | 3.55         | -----            | 4.27         | -----        |
| $R_{exp}$       | -----      | 0.83       | 1.01         | -----            | 1.01         | -----        |
| GOF             | -----      | 2.43       | 3.5          | -----            | 4.21         | -----        |
| $a / 10^{10}m$  | -----      | 2.82(5)    | 2.82(1)      | -----            | 2.816(7)     | -----        |
| $c / 10^{10}m$  | -----      | 14.02(2)   | 14.0(2)      | 13.90(7)         | -----        | -----        |
| $V / 10^{-30}m^3$| -----      | 96.5(5)    | 96(1)        | 95.5(1)          | -----        | -----        |
| $c/a$           | -----      | 4.970(2)   | 4.9521(5)    | 4.9364(4)        | -----        | -----        |

### Discharged electrodes
(after charging to 4.2 V)

| Material        | NMC 801010 | NMC 900505 | NMC 90040501 | NMC 9003.50501.5 | NMC 90030502 | NMC 90050401 |
|-----------------|------------|------------|--------------|------------------|--------------|--------------|
| $R_{wp}$        | 1.22       | 1.31       | 1.84         | 1.26             | 1.57         | 1.64         |
| $R_{exp}$       | 0.81       | 0.82       | 0.83         | 0.83             | 0.82         | 0.85         |
| GOF             | 1.51       | 1.59       | 2.22         | 1.52             | 1.92         | 1.94         |
| $a / 10^{10}m$  | 2.8657(2)  | 2.8714(2)  | 2.8698(2)    | 2.869(6)         | 2.8666(2)    | 2.8739(2)    |
| $c / 10^{10}m$  | 14.231(2)  | 14.217(2)  | 14.218(3)    | 14.22(4)         | 14.235(2)    | 14.209(3)    |
| $V / 10^{-30}m^3$| 101.21(2)  | 101.52(2)  | 101.40(2)    | 101.4(1)         | 101.30(2)    | 101.64(3)    |
| $c/a$           | 4.9660(4)  | 4.9513(5)  | 4.9542(5)    | 4.9566(4)        | 4.9657(4)    | 4.9443(5)    |
Figure S10. a) Lattice parameters $a$ and $c$ changes as a function of Mg content when charging all cells to the same upper cut-off voltage of 4.2 V as determined by Pawley refinements. b) 2nd cycle cell voltage profiles of NMC || graphite cells at 20 °C at 0.1C charging up to 200 mAh g$^{-1}$ without limitation in upper cut-off voltage. c) Lattice parameters $a$ and $c$ evolution when charging all electrodes to the same degree of lithium extraction ratio (200 mAh g$^{-1}$, $x=0.72$, in Li$_{1-x}$TMO$_2$) as determined by Pawley refinements.
Figure S11. Representative Pawley refinements of XRD patterns of pristine Ni-rich Mg-substituted NMC electrodes: a) NMC 801010, b) NMC 900505, c) NMCMg 90040501, d) NMCMg 9003.50501.5 e) NMCMg 90030502 and f) NMCMg 90050401. The panels on the right show expanded views of the (006)/(102) and (018)/(110) reflections slit up as an indication of well-defined layered structure. The black empty dots show the original data points, the solid red lines the calculated results while the blue lines show the differences between the measured and calculated data. R-weighted pattern (Rwp), R-expected (Rexp) and Goodness of fit (GOF) are shown in the inset.
Figure S12. Representative Pawley refinements of XRD patterns of charged (up to 4.2 V) Ni-rich Mg-substituted NMC electrodes: a) NMC 801010, b) NMC 900505, c) NMCMg 90040501, d) NMCMg 9003.50501.5 e) NMCMg 90030502 and f) NMCMg 90050401. The panels on the right show expanded views of the (006)/(102) and (018)/(110) reflections slit up as an indication of well-defined layered structure. The black empty dots show the original data points, the solid red lines the calculated results while the blue lines show the differences between the measured and calculated data. R-weighted pattern ($R_{wp}$), R-expected ($R_{exp}$) and Goodness of fit (GOF) are shown in the inset.
Figure S13. Representative Pawley refinements of XRD patterns of charged (up to 200 mAh g\(^{-1}\); corresponding to \(x\approx0.72\) in Li\(_{1-x}\)TM\(_2\)O\(_2\)) Ni-rich Mg-substituted NMC electrodes: a) NMC 900505, b) NMCMg 90040501, and c) NMCMg 90030502. The black empty dots show the original data points, the solid red lines the calculated results while the blue lines show the differences between the measured and calculated data. R-weighted pattern (\(R_{wp}\)), R-expected (\(R_{exp}\)) and Goodness of fit (GOF) are shown in the inset.
Figure S14. Representative Pawley refinements of XRD patterns of discharged (2.8 V, after charging to 4.2 V) Ni-rich Mg-substituted NMC electrodes: a) NMC 801010, b) NMC 900505, c) NMCMg 90040501, d) NMCMg 9003.50501.5 e) NMCMg 90030502 and f) NMCMg 90050401. The panels on the right show expanded views of the (006)/(102) and (018)/(110) reflections slit up as an indication of well-defined layered structure. The black empty dots show the original data points, the solid red lines the calculated results while the blue lines show the differences between the measured and calculated data. R-weighted pattern ($R_{wp}$), R-expected ($R_{exp}$) and Goodness of fit (GOF) are shown in the inset.
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