Tailoring the Electrochemical Activity of Magnesium Chromium Oxide towards Mg Batteries Through Control of Size and Crystal Structure
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Supplementary Information
Figure S1 Schematics of a) the Continuous Hydrothermal Flow Synthesis (CHFS) apparatus, displaying all key components and b) the Confined Jet Mixer (CJM), displaying the precise mixing arrangement between precursor and supercritical feeds.

Flash Heat Treatment Process

A crucible was inserted into a section of 3/8" pipe, which resided within a larger outer pipe (1") which was securely clamped. The crucible was inserted and removed from the work tube of the tube furnace by sliding the inner pipe within the outer pipe. The outer pipe secured the inner pipe, thereby ensuring the crucible remained inside the work tube without the inner pipe coming into direct contact with the work tube.

Figure S2 – A schematic of the Flash Heat Treatment Process.
Figure S3 TGA/DSC traces of MgCr$_2$O$_{4-x}$(OH)$_{2x}$, where the TGA trace is blue and the DSC trace is black, with a heating ramp of 5 °C min$^{-1}$ from room temperature to 1000 °C. Two endotherms (with minima at ca. 80 °C and 200 °C) were revealed, suggesting two separate water loss processes, i.e. the loss of surface (adsorbed) water and hydroxide to oxide conversion, respectively. Furthermore, a large exotherm with a maximum at ca. 600 °C was observed in the range 200 – 1000 °C and was attributed to crystallization and particle size growth of the MgCr$_2$O$_4$ phase.
Figure S4 Pawley refinements of the MCO samples. (a) MCO (ΔCHFS) powder (b) MCO (ΔBHS) powder (c) MCO (ΔCHFS) electrode pristine (d) MCO (ΔCHFS) electrode charge. Red line: background; Blue crosses: experimental data; green line: calculated intensity; cyan line: between experimental and calculated intensities; blue ticks: positions of Bragg reflections. (a,b) were collected on Bruker D8 Advance X-ray Diffractometer (λ = 1.5406Å), and (c,d) were performed by High resolution X-ray Diffractometer at beamline 11-ID-B of Advanced Photon Source, Argonne National Laboratory (λ = 0.2114 Å). Peaks from carbon black peak and two spikes were excluded in (c,d).
Table S1 Unit cell parameters for different MCO samples, extracted from Pawley refinements

<table>
<thead>
<tr>
<th>Samples</th>
<th>Plots</th>
<th>a (Å)</th>
<th>Vol (Å³)</th>
<th>R_w (%)</th>
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<tbody>
<tr>
<td>MCO (ΔCHFS) Powder</td>
<td>Pristine</td>
<td>8.30411</td>
<td>572.638</td>
<td>1.16%</td>
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<tr>
<td>MCO (ΔBHS) Powder</td>
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<tr>
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<td>573.009</td>
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<tr>
<td>MCO (ΔCHFS) Electrode</td>
<td>Charge</td>
<td>8.29574</td>
<td>570.901</td>
<td>1.65%</td>
</tr>
</tbody>
</table>
Figure S5 N₂ adsorption plots of MCO (ΔCHFS) and MCO (ΔBHS)
Figure S6 EDX for MCO (ΔCHFS) NCs (a) pristine and (b) charge
Figure S7 EDX for MCO (ΔBHS) NCs (a) pristine and (b) charge
Figure S8 Derivative profile of Cr K-edge XANES of (A) MCO (ΔCHFS) and (B) MCO (ΔBHS) electrodes, harvested at pristine, charge, and discharge, as labeled. Data were collected in transmission mode.
Figure S9 Cr L$_{2,3}$-edge XAS of MCO (ΔCHFS) (a) Comparison between pristine and charge (b) Comparison between pristine and discharge