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# Supporting Information

# Low-Cobalt Cathode Materials for High Performance Lithium-ion Battery: Synthesis and Performance Enhancement Methods

Sourav Mallick[1], Arjun Patel[1], Xiao-Guang Sun[2], Mariappan Parans Paranthaman[2], Mingyao Mou[1], Jethrine H. Mugumya[1], Mo Jiang[1], Michael L. Rasche[1], Herman Lopez[3], and Ram B. Gupta*[1]

[1]Department of Chemical and Life Science Engineering, Virginia Commonwealth University, Richmond, VA, 23219, USA.

[2]Chemical Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN, 37831, USA

[3]Ionblox Inc., Fremont, CA, 94538, USA

**Corresponding authors: Email addresses: rbgupta@vcu.edu (Dr. Ram B. Gupta)**

**Table S1.** Electrochemical performance of various low cobalt cathodes with and without modifications

| Material/ Cell configuration | $I_{(003)}/I_{(104)}$ | Voltage (V) | Specific capacity (mAh $g^{-1}$) | Cycling performance | Ref. |
|---|---|---|---|---|---|
| **Without modification** | | | | | |
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| NCM811 | 1.65 | 2.7-4.3 | 203.7 mAh $g^{-1}$ at 0.1 C; 167.2 mAh $g^{-1}$ at 3 C<br><br>(1 C = 200 mAh $g^{-1}$) | 88% capacity retention after 200 cycles at 0.2 C | 1 |
| $LiNi_{0.76}Mn_{0.14}Co_{0.10}O_2$ (calcination temp: 750 ºC)<br><br>(cathode material loading: 12 mg $cm^{-2}$) | 1.26 | 2.7-4.5 (V vs. $Li/Li^+$) | 215 mAh $g^{-1}$ at 0.1 C; 168 mAh $g^{-1}$ at 10 C<br><br>(1 C = 200 mAh $g^{-1}$) | 92% capacity retention after 100 cycles at 1 C | 2 |
| Olive like NCM811 (solvothermal synthesis)<br><br>(cathode material loading: 8.0 mg $cm^{-2}$) | 1.431 | 2.7-4.3 (V vs. $Li/Li^+$) | 193.4 mAh $g^{-1}$ at 0.2 C; >130 mAh $g^{-1}$ at 5 C<br><br>(1 C = 200 mAh $g^{-1}$) | 85.4% capacity retention after 100 cycles at 1 C | 3 |
| Agglomerated NCM811 (solvothermal synthesis)<br><br>(cathode material loading: 4.0 mg $cm^{-2}$) | - | 2.7-4.3 (V vs. $Li/Li^+$) | 203.7 mAh $g^{-1}$ at 0.1 C; 155.6 mAh $g^{-1}$ at 10 C<br><br>(1C = 180 mAh $g^{-1}$) | 86.2% capacity retention after 200 cycles at 1 C | 4 |
| NCM811 (Flame aerosol synthesis)<br><br>(cathode material loading: 46.5 mg $cm^{-2}$) | 0.88 | 2.8-4.3 (V vs. $Li/Li^+$) | 181 mAh $g^{-1}$ at 0.1 C; | 85 % capacity retention after 25 cycles at 0.5 C | 5 |
| **Doping** | | | | | |
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| (Mg-doped) $LiNi_{0.90}Co_{0.02}Mn_{0.05}Mg_{0.02}O_2$/ graphite (full cell)<br><br>(cathode material loading: 12.0 ± 0.5 mg $cm^{-2}$) | - | 2.8-4.2 | 183 ± 1 mAh $g^{-1}$ at 0.1 C<br><br>(1C = 190 mA $g^{-1}$) | 607 cycles to 80% SOH (long-term cycling at 0.33C) | 6 |
| $LiNi_{0.8}Co_{0.07}Fe_{0.03}Mn_{0.1}O_2$ | 1.386 | 2.8-4.3 (V vs. $Li/Li^+$) | 207.5 mAh $g^{-1}$ at 0.1 C; 145.8 mAh $g^{-1}$ at 5 C<br><br>(1C = 180 mAh $g^{-1}$) | 80% SOH after 400 cycles at 0.5 C | 7 |
| $Na^+$ and $Al^{3+}$ dual doped $LiNi_{0.88}Co_{0.08}Mn_{0.04}O_2$<br><br>(cathode material loading: 7.0 mg $cm^{-2}$) | 1.44 | 2.8-4.35 (V vs. $Li/Li^+$) | 212.30 mAh $g^{-1}$ at 0.2 C; 176.46 mAh $g^{-1}$ at 3 C<br><br>(1C = 180 mAh $g^{-1}$) | 84% capacity retention after 50 cycles at 1 C | 8 |

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| Cu-doped NCM811 | 1.70 | 2.8–4.3 V (V vs. $Li/Li^+$) | 226.7 mAh $g^{-1}$ at 0.1 C; 124.1 mAh $g^{-1}$ at 5 C<br><br>(1 C = 200 mAh $g^{-1}$) | >90% capacity retention after 100 cycles is at 1C | 9 |
| Ti-doped NCM811<br><br>(cathode material loading: 2.5 mg $cm^{-2}$) | 1.22 | 2.8-4.6 V (V vs. $Li/Li^+$) | 196 mAh $g^{-1}$ at 0.5 C; 157 mAh $g^{-1}$ at 2 C<br><br>(1 C = 200 mAh $g^{-1}$) | 84% capacity retention after 100 cycles at 1 C | 10 |
| B-doped NCM811<br><br>(cathode material loading: 5 mg $cm^{-2}$) | 1.01 | 3.0-4.3 V (V vs. $Li/Li^+$) | 191 mAh $g^{-1}$ at 0.04 C<br><br>(1 C = 200 mAh $g^{-1}$) | 87% capacity retention after 120 cycles at 0.5 C | 11 |
| **Coating** | | | | | |
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| Tungsten oxide coated NCM811/ graphite (full cell)<br><br>(cathode material loading: 12.0 ± 0.4 mg $cm^{-2}$) | 2.30 | 2.5-4.3 | 184 mAh $g^{-1}$ at 0.1 C; ~75 mAh $g^{-1}$ at 5 C | 800 cycles to 80% SOH | 12 |
| AZO coated NCM811<br><br>(cathode material loading: 1.5-2 mg $cm^{-2}$) | - | 3.0-4.5 (V vs $Li/Li^+$) | 216.1 mAh $g^{-1}$ at 0.1 C; >150 mAh $g^{-1}$ at 10 C | 86.3% capacity retention after 100 cycles at 1 C | 13 |
| $LaPO_4$ coated NCM811 | 1.442 | 3.0-4.3 (V vs $Li/Li^+$) | 196.7 mAh $g^{-1}$ at 0.1 C; 124 mAh $g^{-1}$ at 10 C | 91.2 % capacity retention after 100 cycles at 1 C | 14 |
| $Li_3PO_4$ coated NCM811 | 1.78 | 2.5-4.3 (V vs $Li/Li^+$) | 185.0 mAh $g^{-1}$ at 95 mA $g^{-1}$; ~80 mAh $g^{-1}$ at 950 mA $g^{-1}$ | 96 % capacity retention after 100 cycles at 95 mA $g^{-1}$ | 15 |
| Dihexadecyl phosphate (0.1%)-coated NCM811<br><br>(cathode material loading: 3.0 mg $cm^{-2}$) | - | 2.75-4.3 (V vs $Li/Li^+$) | 204.5 mAh $g^{-1}$ at 0.1 C; 180.1 mAh $g^{-1}$ at 1 C<br><br>(1 C = 200 mAh $g^{-1}$) | 62 % capacity retention after 500 cycles at 1 C | 16 |
| **Dual modification** | | | | | |
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| $Mg^{2+}$ doping and $Li_3PO_4$ coated NCM811 ($MgHPO_4$-modified) | 1.63 | 2.9-4.3 | 203.5 mAh $g^{-1}$ at 0.5 C; 169.4 mAh $g^{-1}$ at 6 C<br><br>(1 C = 200 mAh $g^{-1}$) | 86.3 % capacity retention after 100 cycles at 1 C | 17 |
| Zr doped and Lithium zirconate coated NCM811<br><br>(cathode material loading: 3.8 mg $cm^{-2}$) | 2.18 | 2.8-4.3 (V vs $Li/Li^+$) | 192 mAh $g^{-1}$ at 0.1 C; 100 mAh $g^{-1}$ at 10 C<br><br>(1 C = 200 mAh $g^{-1}$) | 84.3 % capacity retention after 60 cycles at 0.2 C | 18 |
| Mg doping and $Li_3PO_4$ coated $LiNi_{0.91}Co_{0.06}Mn_{0.03}O_2$<br><br>(cathode material loading: 15.5 ± 0.5 mg $cm^{-2}$) | 1.36 | 3.0-4.3 (V vs $Li/Li^+$) | 200 mAh $g^{-1}$ at 0.1 C; 124.6 mAh $g^{-1}$ at 3 C | 73.5 % capacity retention after 80 cycles at 0.1 C | 19 |
| **Core-shell structure** | | | | | |
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| Core-shell structured $[Ni_{0.85}Co_{0.10}Mn_{0.05}]O_2$ (leached with 5 mol% $H_2SO_4$)<br><br>(cathode material loading: 7-8 mg $cm^{-2}$) | 1.04 - 1.11 | 3.0-4.3 (V vs $Li/Li^+$) | ~180 mAh $g^{-1}$ at 0.2 C; ~120 mAh $g^{-1}$ at 20 C | 82.3 % capacity retention after 150 cycles at 0.5 C | 20 |
| NCM811@$Li_2MnO_3$<br><br>(cathode material loading: 3-4 mg $cm^{-2}$) | - | 2.7-4.7 (V vs $Li/Li^+$) | 254 mAh $g^{-1}$ at 20 mA $g^{-1}$<br>233 mAh $g^{-1}$ at 120 mA $g^{-1}$ | 86 % capacity retention after 90 cycles at 120 mA $g^{-1}$ | 21 |
| Core shell NCM811<br>Core: $LiNi_{0.9}Co_{0.05}Mn_{0.05}O_2$<br>Shell: $LiNi_{0.4}Co_{0.03}Mn_{0.3}O_2$ | 1.74 | 2.7-4.6 (V vs $Li/Li^+$) | 213.1 mAh $g^{-1}$ at 0.2 C; 148.6 mAh $g^{-1}$ at 3 C<br><br>(1 C = 180 mAh $g^{-1}$) | 86 % capacity retention after 100 cycles at 1 C | 22 |
| **Concentration gradient** | | | | | |
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| NCM811<br>Ni conc. 84% to 76% from center to edge) | 1.51 | 2.75-4.3 (V vs $Li/Li^+$) | 198.3 mAh $g^{-1}$ at 0.1 C; 182.5 mAh $g^{-1}$ at 1 C | 93.7 % capacity retention after 100 cycles at 5 C | 23 |
| NCM811<br>Ni conc. 84.7% to 67.3% from center to edge) | >1.2 | 2.75-4.3 (V vs $Li/Li^+$) | 201.5 mAh $g^{-1}$ at 0.1 C; 184.2 mAh $g^{-1}$ at 1 C | 98.0 % capacity retention after 200 cycles at 1 C | 24 |
| $TiO_2$-incorporated NCM811 with full conc. Gradient (FCG)<br>Core: $Li[Ni_{0.91}Co_{0.06}Mn_{0.03}]O_2$<br>Surface: $Li[Ni_{0.55}Co_{0.16}Mn_{0.29}]O_2$ | 1.64 | 2.8-4.4 (V vs $Li/Li^+$) | ~180 mAh $g^{-1}$ at 0.1 C; 92 mAh $g^{-1}$ at 10 C<br><br>(0.2 C= 40 mA $g^{-1}$) | 96.0 % capacity retention after 150 cycles at 1 C | 25 |
| Nb-doped and $LiNbO_3$-coated FCG NCM811 | - | 3.0-4.4 (V vs $Li/Li^+$) | 214.8 mAh $g^{-1}$ at 0.1 C; ~162.4 mAh $g^{-1}$ at 10 C<br><br>(1 C = 180 mAh $g^{-1}$) | ~85.1 % capacity retention after 300 cycles at 1 C | 26 |
| Spinel layer on the surface of layered NCM811<br><br>(cathode material loading: 3-4 mg $cm^{-2}$) | 1.450 | 3.0-4.5 (V vs $Li/Li^+$) | 211.2 mAh $g^{-1}$ at 0.1 C; 111.7 mAh $g^{-1}$ at 10 C<br><br>(1 C = 200 mAh $g^{-1}$) | 92.7 % capacity retention after 100 cycles at 1 C | 27 |
| $LiNi_{0.7}Co_{0.13}Mn_{0.17}O_2$ with concentration gradient<br><br>(cathode material loading: 2 mg $cm^{-2}$) | 1.27 | 3.0-4.3 (V vs $Li/Li^+$) | 187.8 mAh $g^{-1}$ at 0.1 C; 111.7 mAh $g^{-1}$ at 10 C<br><br>(0.1 C = 20 mA $g^{-1}$) | 86.5 % capacity retention after 300 cycles at 1 C | 28 |
| **Single crystal structure** | | | | | |
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| NCM811<br>(cathode material loading: ~3 mg $cm^{-2}$) | 1.27 | 3.0-4.3 (V vs $Li/Li^+$) | ~200 mAh $g^{-1}$ at 20 ma $g^{-1}$ | 82 % capacity retention after 50 cycles at 20 mA $g^{-1}$ | 29 |

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| NCM811<br>(cathode material loading: ~3 mg cm$^{-2}$) | - | 2.8-4.3<br>(V vs Li/Li$^{+}$) | 197.9 mAh g$^{-1}$ at 0.2 C;<br>161.8 mAh g$^{-1}$ at 1 C<br><br>(1 C = 170 mAh g$^{-1}$) | 79.7 % capacity retention after 200 cycles at 1 C | 30 |
| Oct-SC811 [predominating (012)-plane]<br><br>Poly-SC811 [predominating (104)-plane]<br><br>(cathode material loading: ~6 mg cm$^{-2}$) | - | 3.0-4.3<br>(V vs Li/Li$^{+}$) | ~200 mAh g$^{-1}$ at 0.1 C;<br><165 mAh g$^{-1}$ at 10 C<br><br>~200 mAh g$^{-1}$ at 0.1 C;<br>~165 mAh g$^{-1}$ at 10 C | 15.7 % capacity loss after 100 cycles at 6 C<br><br>5.9 % capacity loss after 100 cycles at 6 C | 31 |
| NCM811 (lithium excess 50%) | 1.669 | 2.8-4.3<br>(V vs Li/Li$^{+}$) | 226.9 mAh g$^{-1}$ at 0.1 C;<br>140 mAh g$^{-1}$ at 5 C<br><br>(1 C = 200 mAh g$^{-1}$) | 74 % capacity retention after 200 cycles at 5 C | 32 |
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| **Electrolyte Additives** | | | | | |
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| NCM811<br><br>**Without additive**<br><br>**With Additive:** Lithium difluorooxalate borate (2 % LiODFB) | - | 3.0-4.3<br>(V vs Li/Li$^{+}$) | <200 mAh g$^{-1}$ at 0.1 C;<br><100 mAh g$^{-1}$ at 4 C<br><br>>200 mAh g$^{-1}$ at 0.1 C;<br>>120 mAh g$^{-1}$ at 4 C | 68.6 % capacity retention after 50 cycles at 0.2 C<br><br>85 % capacity retention after 50 cycles at 0.2 C | 33 |
| NCM811//Graphite (full cell)<br><br>**Without additive**<br><br>**With Additive:** 1 vol. % tris(trimethylsilyl)phosphite (TMSPi) + vinylene carbonate (VC)<br><br>(cathode material loading: 10.5-11.5 mg cm$^{-2}$) | - | 3.0-4.3 V | 180 mAh g$^{-1}$ at C/20;<br>>130 mAh g$^{-1}$ at C/3<br><br>>190 mAh g$^{-1}$ at C/20;<br>~170 mAh g$^{-1}$ at C/3<br><br>(1 C = 190 mAh g$^{-1}$) | 91 % capacity retention after 200 cycles at C/3<br><br>99.95 % capacity retention after 200 cycles at C/3 | 34 |
| NCM811//Silicon-Graphite (full cell)<br><br>**Without additive**<br><br>**With Additive:** 2 wt. % tris(trimethylsilyl)phosphite (TMSPi) | - | 2.7-4.4 V | ~200 mAh g$^{-1}$ at 0.1 C;<br>~125 mAh g$^{-1}$ at 2C<br><br>~200 mAh g$^{-1}$ at 0.1 C;<br>>125 mAh g$^{-1}$ at 2C | 55 % capacity retention after 50 cycles at 0.5<br><br>80 % capacity retention after 50cycles at 0.5 | 35 |

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| (cathode material loading: 10 mg $cm^{-2}$) | | | | | |

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