Supporting Information for

Mitigating the capacity and voltage decay of lithium-rich layered oxide cathodes by fabricating Ni/Mn graded surface

Feng Li, a,b Yangyang Wang, b Shilun Gao, c Peiyu Hou a and Lianqi Zhang c

a School of Physics and Technology, University of Jinan, Jinan, 250022, Shandong Province, China.
b School of Materials Science and Engineering, National Institute for Advanced Materials, Nankai University, Tianjin, 300350, China.
c Tianjin Key Laboratory for Photoelectric Materials and Devices, School of Materials Science and Engineering, Tianjin University of Technology, Tianjin, 300384, China.

*E-mail: sps_houpy@ujn.edu.cn (P. Hou)
*E-mail: zhanglianqi@gmail.com (L. Zhang)
Figure S1 Schematic illustration of preparing the aimed Ni/Mn graded precursors and lithium-rich layered oxides via a developed co-precipitation route and solid-state reactions.

Figure S2 The growth mechanism of Ni/Mn graded micron-sized spherical secondary particles as precursors during co-precipitation process in the CSTR.
Figure S3 (a-c) SEM images and (d) particle size distribution of the normal precursors [Ni$_{0.183}$Co$_{0.167}$Mn$_{0.65}$]CO$_3$.

Figure S4 SEM images of the normal LLOs.
Figure S5 XPS of the (a,b) Ni 2p<sub>3/2</sub>, (c,d) Co 2p<sub>3/2</sub>, (e,f) Mn 2p<sub>3/2</sub> and (g,h) C 1s for the both pristine LLOs.
Figure S6 (a) The initial charge/discharge curves and (b) the cycling stability of the Ni/Mn graded LLO electrodes at high working temperature of 50 ℃.

Figure S7 XPS of the C1s for both the cycled LLO electrodes in the fully discharged state after 200 cycles at a rate of 0.5C and 25 ℃.

Table S1 The molar ratio of Ni/Co/Mn by ICP–AES for both as-prepared precursors.

<table>
<thead>
<tr>
<th>Samples</th>
<th>Ni</th>
<th>Co</th>
<th>Mn</th>
</tr>
</thead>
<tbody>
<tr>
<td>Normal precursors</td>
<td>0.183</td>
<td>0.167</td>
<td>0.650</td>
</tr>
<tr>
<td>Ni/Mn graded precursors</td>
<td>0.184</td>
<td>0.168</td>
<td>0.648</td>
</tr>
</tbody>
</table>