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Determination of the structure of cobalt-free Li-Mn-rich oxides

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Cobalt-free lithium- and manganese-rich layered oxides (Li[LixNiyMn1-x-y]O2, LMLOs) has catalyzed intensive research efforts to determine their structure that could accommodate a relatively large amount of lithium ions. This feature can make the LMLO electrodes more competitive than the conventional Li[NixCoyMn1-xy]O2 (NCM) cathodes for Li-ion batteries. However, whether LMLOs should be regarded as accumulation of layered monoclinic phase (C2/m) and layered rhombohedral phase (R-3m) nanodomains or as a layered monoclinic single-phase solid solution (C2/m) remains an open question. Herein, high-resolution neutron powder diffraction at the instrument SPODI was used to investigate the localisation and quantification of lithium and oxygen in the structure. Combined with the analysis of synchrotron radiation diffraction and electron diffraction, we demonstrate that the structure of Li[Li0.2Ni0.2Mn0.6]O2 is a single monoclinic solid solution layered structure with ultrathin spinel/rock-salt-type surface. These results contribute to a profound analysis of the relationship between electrochemical performances and the structure of LMLOs.

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