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Nanoscale interactions governing the performance of lithium- and manganese-rich layered oxides cathodes

Lithium-rich, transition-metal-oxide cathodes are among the most promising materials for the next lithium-ion batteries generation because they operate at high voltages and deliver high capacities. However, their cycle life remains limited and individual roles of the transition-metals are still not fully understood, limiting the applications. We exploit bulk-sensitive X-ray absorption and emission spectroscopy on $\text{Li}[\text{Li}_{0.2}\text{Ni}_{0.16}\text{Mn}_{0.56}\text{Co}_{0.08}]\text{O}_2$ to inspect the behavior of Mn, which is generally considered inert upon the electrochemical process. During the first charge Mn appears to be redox active showing a partial transformation from high-spin Mn^{4+} to Mn^{3+} in both high and low spin configurations, where the latter is expected to favor reversible cycling. The Mn redox state along cycling continues changing in opposition to the expected charge compensation and is correlated with Ni oxidation/reduction, also spatially. The findings suggest that the strain induced on the Mn-O sublattice by the Ni oxidation is triggering the Mn reduction. These results unravel the role of Mn in controlling the electrochemistry of this family of compounds, which should be taken into account when designing new cathodes.