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Thermally induced cation re-ordering in activated Li- and Mn-rich layered oxide Li-ion cathode materials

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Layered transition metal oxides such as the 'Li- and Mn-rich' layered oxides, formed as the composites between Li_2MnO_3 and LiMO_2 ($M = \text{Ni, Co, Mn}$) (NCM) are promising candidates for next generation Li-ion battery cathodes that offer high reversible capacities ($> 250 \text{ mAh/g}$) and higher safety together with reduced costs. For an economic and efficient operation as cathode materials in electrified vehicles for example, knowledge about mechanisms of degradation in order to optimize these materials for longer lifetime is mandatory.

So far, a gradual decrease of the energy density during electrochemical cycling called 'voltage and capacity fade' is a major drawback of this material class. A local cation rearrangement towards a cubic LiMn_2O_4 spinel-like symmetry during de-/lithiation is supposed to cause this energy decay in many publications.

In order to study the thermodynamically preferred cation arrangement in dependency of the lithiation grade, the structural transformation was thermally induced and investigated by global synchrotron diffraction as well as local nuclear magnetic resonance and X-ray absorption techniques. Furthermore, the structural characteristics of the artificially fatigued samples were compared with conventional highly cycled electrodes and finally correlated with their electrochemical properties.

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