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Mechanistic and structural investigation of Li_xMnO₂ cathodes during cycling in Li-ion batteries

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Increasingly there is demand for clean energy sources and suitable batteries to store this energy. Manganese dioxide and lithiated variants are a promising alternative to conventional Li-ion cathodes due to their cost, abundance, safety and electrochemical performance. Cathodes which operate by a single-phase lithium insertion/extraction process can offer some intrinsic advantages over those with two-phase processes. In this work, in-situ and ex-situ synchrotron X-ray diffraction (XRD) is used to investigate the structural evolution and lithium insertion/extraction mechanism of various LixMnO2 cathodes that have been derived from γ-MnO2. Li0.30MnO2 is found to cycle solely with a single-phase mechanism, in contrast to previous literature reports, with only subtle changes in the crystal structure. However, a better cycling discharge capacity is realised through a two-step lithiation synthesis, thermally lithiated Li0.08MnO2 which is then electrochemically lithiated to Li0.33MnO2. After an irreversible two-phase reaction early in the first discharge, this material cycles by a single-phase reaction with good structural reversibility and a stable unoptimised cycling capacity of 120 mAh/g. Comparing cathodes using a combination of in-situ and ex-situ synchrotron XRD data allows us to rationalise cathodic performance with structure and thereby directing research into promising candidates.

Keywords or phrases (comma separated)

Manganese dioxide, in-situ synchrotron XRD, Li-ion, energy storage

Summary

Primary author(s) : Mr DOSE, Wesley (University of Newcastle)

Co-author(s) : SHARMA, Neeraj (UNSW); Prof. DONNE, Scott (University of Newcastle)

Presenter(s) : Mr DOSE, Wesley (University of Newcastle)

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