

Investigation on Capacity Fading Mechanism of LiNiO₂ Cycled above 4.2 V

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Among the existing cathodes for LIBs, the LiNiO₂ (LNO) layered oxide, isostructural with LiCoO₂ (LCO), has been considered as an alternative cathode because of its high reversible capacity, good rate capability, and relatively low material cost compared to LCO. The requirement for significant improvement in energy density of the LIBs for EVs has recently rekindled the interest in high-capacity LNO.^[1] However, LiNiO₂ cathode has several issues; especially LNO suffered from multiphase transition during cycling.^[2] To explicitly demonstrate the effect of the phase transitions during Li deintercalation/intercalation on the Li-ion intercalation stability of LiNiO₂, spherical stoichiometric LiNiO₂ particle was prepared and cycled at different cutoff voltages. The capacity retention was greatly improved by suppressing the H2 → H3 phase transition at 4.1 V such that 95 % of the initial capacity (164 mAh g⁻¹) was retained after 100 cycles when cycled at 4.1 V. At 4.2 and 4.3 V, continuous capacity loss (81 % of 191 mAh g⁻¹ at 4.2 V and 75 % of 232 mAh g⁻¹ at 4.3 V after 100 cycles) was observed during cycling and these electrodes incurred extensive structural damages from the repeated lattice contraction and expansion accompanying the H2 → H3 transition. This repeated structural damage gave rise to micro-, hairline and nano-scale cracks which were observed by transmission electron microscopy, in agreement with the cycling data.^[3] To improve the cycling stability of these extremely Ni-rich LiNiO₂ cathodes, further developments in the particle morphology to dissipate the intrinsic lattice strain, surface stabilization, and composition modification are required to attain a satisfactory long-term cycling stability, and hence battery life for EVs.

References:

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