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Improvement of the electrochemical performance of spinel $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ by stabilization of the electrode/electrolyte interfaces with the electrolyte additive

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Abstract. High-voltage spinel $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ cathode results in 4.7 V Li-ion batteries with a high power capability, but the electrochemical performances are limited by electrode/electrolyte interfacial reactivity at high potential. We demonstrate here that using a new electrolyte additive based on LiFSI, $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4//\text{Li}$ cells upon cycling show reduced capacity fading and improved coulombic efficiency. The capacity retention after 200th cycle is ~96% using the electrolyte additive, compared with ~89% without the electrolyte additive. The coulombic efficiency for cells with the electrolyte additive shows an increase from ~96.5% to ~98.5%. Detailed X-ray photoelectron spectroscopy and electrochemical impedance spectroscopy measurements reveal that LiPF_6 degradation during cycling via formation of a passivation film at the surface of the electrode. Our results further reveals that the surface film is composed of organic compounds resulting from degradation of the electrolyte additive. The formed surface film is much thicker (~5 nm) but less resistive on the electrode cycled with the electrolyte additive, compared with that without the electrolyte additive.

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