Investigation of Lithium difluorophosphate as a novel salt-type Electrolyte Additive for LiNi$_{0.5}$Mn$_{0.25}$Co$_{0.25}$O$_2$ cathodes

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Although the LiNi$_{0.5}$Mn$_{0.25}$Co$_{0.25}$O$_2$ holds the merits of high theoretical capacities and a relatively high operating voltage, the battery performance suffers from the severe cycling decay due to the unstable solid electrolyte interface on the cathode. Therefore, improving the battery performance via stabilizing the electrode/electrolyte interface is a main focus of the additive research [1]

There are some attractive examples in literature which describe electrolyte salt as film-forming additive[2]. In this work, we present LiPO$_2$F$_2$ as a salt-type additive to enhance the cycling stability of large-size crystallite LiNi$_{0.5}$Mn$_{0.25}$Co$_{0.25}$O$_2$ cathodes. Results demonstrate that addition of LiPO$_2$F$_2$ widening the stability of the electrolyte. When the content of LiPO$_2$F$_2$ increased to 1.0 wt%, the capacity remains 156 mAh g$^{-1}$ with an efficiency of 91.6% after 100 cycles at 55 ºC. The cathode with 1 wt% LiPO$_2$F$_2$–added electrolyte formed a relatively thin (15 - 20 nm) and uniform SEI layer, which mean LiPO$_2$F$_2$ has a significantly positive effect on internal resistances (R$_{SEI}$ and R$_{CT}$) and mitigate undesirable electrolyte decomposition. We also observed that the concentration of O-P-F species decreases on the XPS spectra of P2p, indicating a less extensive decomposition of LiPO$_2$F$_2$ formed LiF and Li$_3$PO$_4$ during the cycling.

Figure 1. Linear sweep voltammetry (LSV) scans of electrolyte with various amount of LiPO$_2$F$_2$(a), The specific capacity of the LiNi$_{0.5}$Mn$_{0.25}$Co$_{0.25}$O$_2$/Li cells as a function of the cycle number at various amount of LiPO$_2$F$_2$ at 55 ºC (3 cycles of formation process at a rate of C/10 and cycling at a rate of 1C)(b), TEM images with 1 wt% LiPO$_2$F$_2$ electrolyte after 200th cycles(c), Nyquist plot of LiNi$_{0.5}$Mn$_{0.25}$Co$_{0.25}$O$_2$/Li cells without and with 1.0 wt% LiPO$_2$F$_2$ additive (d), XPS spectra of P2p with 1.0 wt% LiPO$_2$F$_2$-containing electrolytes(e).

References: