Indole derivatives as film forming additives for high-voltage lithium ion positive electrode

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For the last decades, lithium-ion batteries (LIBs) have been used as the power source for mobile electronic devices [1]. In the near future, the market of LIBs seems to expand to the power sources for electric vehicles and energy storage devices [2]. The existing LIB technologies, however, have serious performance limitation on aspect of energy density as well as safety and cycle life for their new applications. As an effort to increase the energy density of LIBs, a few high-voltage positive electrodes have been tested. One of the spotlighted materials is the lithium-excess layered oxides (also named over-lithiated layered oxides, OLOs), $Li_xMn_yCo_zNi_aO_2$, which have a higher working voltage [3]. The high working voltage must be beneficial for the energy density of LIBs, but poses a serious problem on the electrolyte stability against oxidation, which is even more serious at elevated temperatures. One way to solve or mitigate this problem must be an addition of film-forming agents into electrolyte solution, which decompose to form a protective layer on OLO surface to prevent further electrolyte decomposition [4-7].

In this study, we found that cathode surface modification using N-heterocyclic aromatic compounds (indole derivatives) exerted a significant influence on battery performance. To test the feasibility for a film-forming agent, their oxidation potentials are calculated by using the density functional theory (DFT) and compared with those of the carbonate solvents. Superior cycle performance indicated that stable films could be formed on the cathode surface by using these additives. These additives could also enhance the cycle performance of $LiCoO_2$/graphite cylindrical cell.

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