Improved performance of $LiNi_{0.5}Mn_{1.5}O_4$ spinel with electrolytes containing 1,3,5-Trihydroxybenzene

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In the last years, lithium-ion batteries have been used as the power source for mobile devices [1]. In the near future, it is expected that the market of lithium-ion batteries will expand for applications in electric vehicles and energy storage devices. However, there is some serious performance limitation concerning the energy density of such batteries as well as safety issues for their new applications.

In order to increase the energy density of lithium-ion batteries, high-voltage positive electrodes have been tested. $LiMn_2O_4$ is one of the most promising cathode materials due to abundance and nontoxicity, but it shows lower discharge specific capacity compared to $LiCoO_2$ and a poor cycling behavior at elevated temperature. To reach higher energy density lithium ion batteries, there is growing interest in high voltage cathode materials that operate at potential more positive than 4.5 V vs Li and exhibit higher energy densities. Transition metal-substituted spinel materials ($LiM_xMn_{2-x}O_4$, M = Co, Ni) showed a higher voltage plateau originating from oxidation of substituted transition metals (M), and delivered an higher capacity. The high working voltage of $LiNi_{0.5}Mn_{1.5}O_4$ must be beneficial for the energy density but raises a problem on the stability of electrolyte in contact with the cathode surface at high operating voltages, and especially at elevated temperatures.

Surface coating with various inorganic oxides has been achieved with success to inhibit the sides reactions of the $LiNi_{0.5}Mn_{1.5}O_4$ with electrolyte [2]. The surface coated cathodes have superior cyclability but the surface-coating method has in some case a negative effect on the discharge capacity of the cathode.

Another way to improve the performance is the incorporation of film additives to form a protective layer on $LiNi_{0.5}Mn_{1.5}O_4$ surface to prevent further electrolyte decomposition on the positive electrode. The improvement of novel electrolytes capable of long-term reversible cycling to high voltage is of great interest. Few studies on the formation of a protective film on positive electrodes are published. In this study, 1,3,5-trihydroxybenzene (phloroglucinol, THB hereafter) which is known to undergo oxidative polymerization has been chosen for the good properties of the resulting films [3 4]. Formation and stability of the THB-based film was examined by galvanostatic cycling and surface analysis.

The electrochemical cells were cycled in the potential range 3.5 to 5 V vs. Li. The cell with 0.5% THB in the electrolyte shows higher discharge capacity over 90 cycles presumably because of the higher Coulombic efficiency of the cell cycled with the electrolyte containing 0.5% THB. The cells with THB added electrolyte have superior Coulombic efficiency (Figure 1) relative to the cells containing no THB after 90 cycles, with values of 98.8, 98.7, and 96.5 %, for 0.5% THB, 1% THB, and standard electrolyte, respectively.

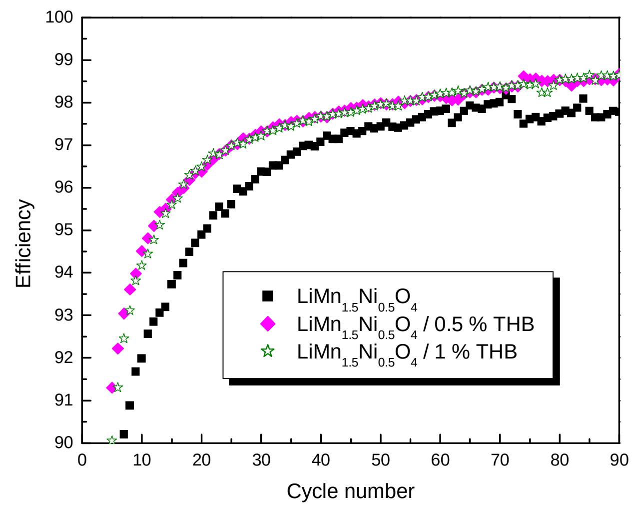

Figure 1: Coulombic efficiency of Li/$LiMn_{1.5}Ni_{0.5}O_4$ cells with standard and THB containing electrolyte

References

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