

Polyimide Gel Polymer Electrolyte-Directed Nanoscale Wrapping of High-Voltage $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$ Cathode Active Materials for Lithium-Ion Batteries

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A new and facile approach for the surface modification of high-voltage $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$ cathode active materials is demonstrated. This strategy is based on polyimide (PI) gel polymer electrolyte (GPE)-directed nanoscale wrapping. The PI coating layer successfully wraps a large area of the $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$ surface via thermal imidization of (pyromellitic dianhydride/oxydianiline) polyamic acid. Salient features of the PI wrapping layer are the highly-continuous surface coverage with nanometer thickness (~ 10 nm) and the facile ion transport through the nanoscale PI layer. Based on a sound understanding of the nanoarchitected PI wrapping layer, its influence on the cell performance and thermal stability of high-voltage $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$ is investigated as a function of charge cut-off voltage (herein, 4.6 and 4.8 V). The anomalous PI wrapping layer substantially improves the high-voltage cycling performance and alleviates the interfacial exothermic reaction between delithiated $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$ and liquid electrolyte. These results demonstrate that the PI wrapping layer effectively prevents the direct exposure of $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$ surface to liquid electrolytes that are highly vulnerable to electrochemical decomposition at high charge voltage conditions, thus behaving as a novel ion-conductive protection skin that mitigates the unwanted interfacial side reactions.

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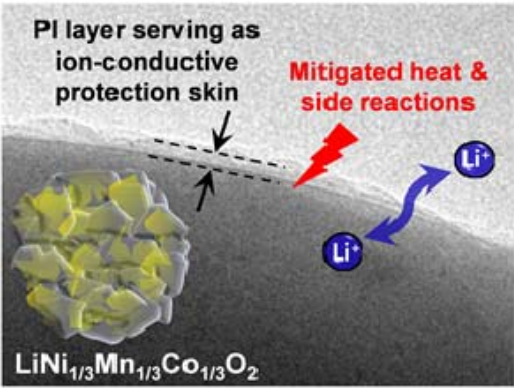


Fig. 1. A TEM photograph of PI-wrapped $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$. The insets indicate schematic illustrations of nanoarchitected PI wrapping layer formed onto $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$ and its function as an ion-conductive protection skin to suppress the unwanted interfacial side reactions.

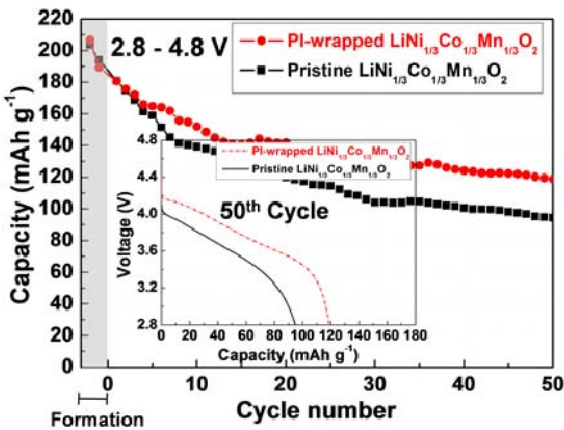


Fig. 2. Discharge capacities (charge/discharge current density = 1.0 C/1.0 C) as a function of cycle number for cells assembled with pristine or PI-wrapped $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$ under a voltage range between 2.8 - 4.8 V. The inset indicate the discharge profiles of cells after the 50th cycle at a given voltage range.