

Synergy Surface Modification for High Performance Li-Rich Cathode

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Abstract : The growing grievous environment problems together with the exhaustion of energy resources put urgent demands for developing high energy density. Considering the factors including capacity, resource and environment, Manganese-based lithium-rich layer-structured cathode materials $x\text{Li}_2\text{MnO}_3 \cdot (1-x)\text{LiMO}_2$ ($M = \text{Ni, Co, Mn, and other metals}$) are drawing increasing attention due to their high reversible capacities, high discharge potentials, and low cost. They are expected to be one type of the most promising cathode materials for the next-generation Li-ion batteries (LIBs) with higher energy densities. Unfortunately, their commercial applications are hindered with crucial drawbacks such as poor rate performance, limited cycle life and continuous falling of the discharge potential. With decades of extensive studies, significant achievements have been obtained in improving their cyclability and rate performances, but they cannot meet the requirement of commercial utilization till now. One major problem for lithium-rich layer-structured cathode materials (LLOs) is the side reaction during cycling, which leads to severe surface degradation. In this process, the metal ions can dissolve in the electrolyte, and the surface phase change can hinder the intercalation/deintercalation of Li ions and resulting in low capacity retention and low working voltage. To optimize the LLOs cathode material, the surface coating is an efficient method. Considering the price and stability, Al_2O_3 was used as a coating material in the research. Meanwhile, due to the low initial Coulombic efficiency (ICE), the pristine LLOs was pretreated by KMnO_4 to increase the ICE. The precursor was prepared by a facile coprecipitation method. The as-prepared precursor was then thoroughly mixed with Li_2CO_3 and calcined in air at 500°C for 5h and 900°C for 12h to produce $\text{Li}_{1.2}[\text{Ni}_{0.2}\text{Mn}_{0.6}]\text{O}_2$ (LNMO). The LNMO was then put into 0.1ml/g KMnO_4 solution stirring for 3h. The resultant was filtered and washed with water, and dried in an oven. The LLOs obtained was dispersed in $\text{Al}(\text{NO}_3)_3$ solution. The mixture was lyophilized to confer the $\text{Al}(\text{NO}_3)_3$ was uniformly coated on LLOs. After lyophilization, the LLOs was calcined at 500°C for 3h to obtain LNMO@LMO@ALO. The working electrodes were prepared by casting the mixture of active material, acetylene black, and binder (polyvinylidene fluoride) dissolved in N-methyl-2-pyrrolidone with a mass ratio of 80: 15: 5 onto an aluminum foil. The electrochemical performance tests showed that the multiple surface modified materials had a higher initial Coulombic efficiency (84%) and better capacity retention (91% after 100 cycles) compared with that of pristine LNMO (76% and 80%, respectively). The modified material suggests that the KMnO_4 pretreat and Al_2O_3 coating can increase the ICE and cycling stability.

Keywords : Li-rich materials, surface coating, lithium ion batteries, Al_2O_3

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