Enhancing the Structural and Electrochemical Performance of Li-Rich Layered Metal Oxides Cathodes for Li-Ion Battery by Coating with the Active Material

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Abstract : The Li-rich layered metal oxides (LLO) are the most promising candidates for promising electrodes of high energy Li-ion battery (LIB). In literature, these electrode system has either been designed as a hetero-structure of the primary components (composite) or as a core-shell structure with improved electrochemistry reported for both configurations when compared with its primary components. With the on-going efforts to improve on the electrochemical performance of the LIB, it is important to investigate comparatively the structural and electrochemical characteristics of the core-shell like and 'composite' forms of these materials with the same compositions and synthesis conditions which could influence future engineering of these materials. Therefore, this study concerns the structural and electrochemical properties of the 'composite' and core-shell like LLO cathode materials with the same nominal composition of 0.5Li2MnO3-0.5LiNi0.5Mn0.3Co0.2O2 $(LiNi_{0.5}Mn_{0.3}Co_{0.2}O_2 as core and Li_2MnO_3 as the shell)$. The results show that the core-shell sample (-CS) gave better electrochemical performance than the 'composite' sample (-C). Both samples gave the same initial charge capacity of ~300 mAh/g when cycled at 10 mA/g and comparable charge capacity (246 mAh/g for the -CS sample and 240 mAh/g for the -C sample) when cycled at 200 mA/g. However, the -CS sample gave a higher initial discharge capacity at both current densities. The discharge capacity of the -CS sample was 232 mAh/g and 164 mAh/g while the -C sample is 208 mAh/g and 143 mAh/g at the current densities of 10 mA/q and 200 mA/q, respectively. Electrochemical impedance spectroscopy (EIS) results show that the -CS sample generally exhibited a smaller resistance than the -C sample both for the uncycled and after 50th cycle. Detailed structural analysis is on-going, but preliminary results show that the -CS sample had bigger unit cell volume and a higher degree of cation mixing. The thermal stability of the -CS sample was higher than the -C sample. XPS investigation also showed that the pristine -C sample gave a more reactive surface (showing formation of carbonate species to a greater degree) which could result in the greater resistance seen in the EIS result. To reinforce the results obtained for the 0.5Li₂MnO₃-0.5LiNi_{0.5}Mn_{0.3}Co_{0.3}O₂ composition, the same investigations were extended to another 'composite' and core-shell like LLO cathode materials also with the same nominal composition of 0.5Li₂MnO₃-0.5LiNi_{0.3}Mn_{0.3}Co_{0.3}O₂. In this case, the aim was to determine the electrochemical performance of the material using a low Ni content (LiNio.3Mno.3Coo.3O2) as the core to clarify the contributions of the core-shell configuration to the electrochemical performance of these materials. Ni-rich layered oxides show active catalytic surface leading to electrolyte oxidation resulting in poor thermal stability and cycle life. Here, the core-shell sample also gave better electrochemical performance than the 'composite' sample with 0.5Li₂MnO₃-0.5LiNi_{0.3}Mn_{0.3}Co_{0.3}O₂ composition. Furthermore, superior electrochemical performance was also recorded for the core-shell like spinel modified LLO (0.5Li2MnO₃-0.45LiNio.5Mno.3Coo.2O₂-0.05LiNio.5Mn1.5O₄) when compared to the composite system. These results show that the core-shell configuration can generally be used to improve the structural and electrochemical properties of the LLO and spinel modified LLO materials.

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