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Architectural Variations of $\text{Li}_2\text{MnO}_3\text{-Li}_{0.69}\text{MnO}_2$ Core-Shell Cathodes During the Cycling Process

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The core-shell architecture has garnered significant interest for electrode materials owing to the ability to enhance conductivity, stability and enhanced surface functionality. Due to its high capacity and energy density, the O₃-type Li_2MnO_3 layered cathode material is a potential candidate electrode for large-scale energy storage. However, it tends to undergo structural transformation from layered to spinel configuration during charge cycles due to irreversible oxygen loss. Recent advances in surface coating techniques have improved the electrochemical performance of these cathodes by enhancing conductivity, stabilizing structures, and preventing harmful reactions with the electrolyte. One of the setbacks with this strategy is the delamination of the core from the rigid shell attributed to the radial tensile radial stress on the interface of the core shell. This compromises the mechanical integrity of the core-shell structure and affects the electrochemical performance of LIBs. In this study, Li_2MnO_3 is coated with $\text{Li}_{0.69}\text{MnO}_2$ for the first time, a layered material known for its stability (no phase transformation) and high ionic conductivity. Molecular dynamics simulations, using the DL_POLY code, were employed to examine the cycling performance of the $\text{Li}_2\text{MnO}_3\text{-Li}_{0.69}\text{MnO}_2$ core-shell system by delithiated from Li_2MnO_3 to LiMnO_3 and to closely monitoring the potential risk of delamination under various temperature conditions. The simulations were carried out using the Nose-Hoover thermostat under the NVT ensemble with temperatures ranging from 300 K to 1500 K. The structural snapshots obtained indicated a fluctuating pattern in the system's structural stability at different temperatures. Notably, at certain temperatures, the core-shell system lost a significant number of atoms from both the core and shell, while at other temperatures, the system regained order with minimal atom loss. Additionally, the lithium diffusion coefficients varied with lithium concentration, showing higher diffusion values for $\text{Li}_{1.3}\text{MnO}_3$ and $\text{Li}_{1.7}\text{MnO}_3$ at lower temperatures and $\text{Li}_{1.4}\text{MnO}_3$ demonstrated highest diffusion rates of 1.25 nm²/s at 1500 K whilst the highest attainable diffusion was that of $\text{Li}_{1.7}\text{MnO}_3$, 3.02 nm²/s at 1200 K. Overall, temperatures where greater atom loss occurred show high diffusion though this trend was not consistent across all concentrations. The simulations were conducted using 48 cores and 5 nodes.

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