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Morphology Control to Enable High Capacity Li-Rich Disordered Rock Salt Cathodes

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Abstract

Li-rich disordered rock salt (DRS) oxides are a promising class of cathode materials with the wide chemical space to be explored. The high capacity (>300mAh/g) of this class of material can be explained by the reversible redox chemistry of the oxide anions, which sets it apart from the conventional layered cathode materials that rely only on the transition metal redox. However, these materials suffer from poor ionic and electronic transport properties: Most previous studies report electrochemical performance at low current rate and elevated temperature. Even then, the particle size needs to be reduced to sub-micrometer size, often by high-energy ball milling, to get reasonable capacities.

To mitigate this issue, we performed a detailed study of the synthesis of three different Nb-based Li-rich DRS materials - Li_3NbO_4, $Li_{1.3}Fe_{0.4}Nb_{0.3}O_2$, and $Li_{1.3}Mn_{0.4}Nb_{0.3}O_2$. Systematic evaluation shows that both the synthesis conditions and the reagents used have a large effect on the phase and morphology of the material synthetized, and therefore on its electrochemical performance. Without varying the synthesis method, the extent of cation ordering, the particle morphology, and the degree of elemental segregation can be controlled by a careful choice of the metal oxide precursors. This study helps the community to distinguish important synthesis criteria in order to design Li-rich DRS cathode materials with improved electrochemical performance.