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A Truncated Manganese Spinel Cathode for Excellent Power and Lifetime in Lithium-ion Batteries

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Spinel-structured lithium manganese oxide (LiMn_2O_4) cathodes have been successfully commercialized for various lithium battery applications and are among the strongest candidates for emerging large-scale applications¹. Despite its various advantages including high power capability, however, LiMn_2O_4 chronically suffers from limited cycle life, originating from well-known Mn dissolution^{2,3}. An ironical feature with the Mn dissolution is that the surface orientations supporting Li diffusion and thus the power performance are especially vulnerable to the Mn dissolution, making both high power and long lifetime very difficult to achieve simultaneously⁴. In this investigation⁵, we address this contradictory issue of LiMn_2O_4 by developing a truncated octahedral structure in which most surfaces are aligned to the crystalline orientations with minimal Mn dissolution while a small portion of the structure is truncated along the orientations to support Li diffusion and thus facilitate high discharge rate capabilities⁵. When compared to control structures with much smaller dimensions, the truncated octahedral structure as large as 500 nm exhibits better performance in both discharge rate performance and cycle life, thus resolving the previously conflicting aspects of LiMn_2O_4 .

For this research, the morphology of LiMn_2O_4 was controlled by Mn_3O_4 or $\text{Mn}(\text{OH})_2$ prior to their lithiation through solid state reactions with Li precursors to complete the final structures with the appropriate stoichiometry. See schematics for the truncated octahedral and other structures in Fig. 1. The morphologies and crystal structures were verified by SEM, TEM, and XRD characterizations.

Electrochemical tests carried out for the three structures shown in Fig. 1, the truncated octahedral structure (Oh^T) exhibits far superior performance in power and cycling properties compared to those of the other control samples (Fig. 2). The better performance is attributed to its unique structure where severe Mn dissolution is suppressed by the dominant $\{111\}$ surfaces while Li ions diffuse efficiently along the truncated $\{110\}$ surfaces. The other control samples lack such dual surface orientations and thus exhibit worse performance in cycle life and/or rate capability.

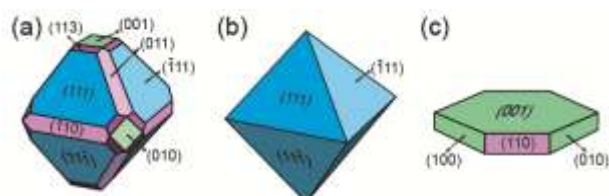


Fig. 1. Schematic illustrations of LiMn_2O_4 with (a) truncated octahedral, (b) bare octahedral, and (c) platelet structures.

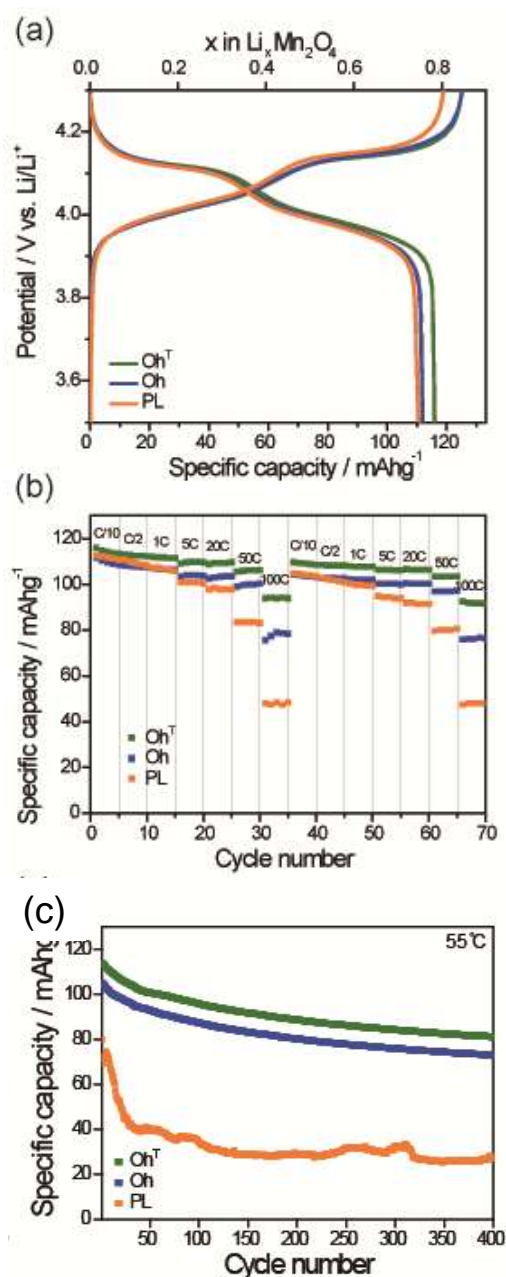


Fig. 2. (a) First potential profiles, (b) rate capabilities, and (c) cycle lives at 55 °C of truncated octahedral (Oh^T), octahedral (Oh), and platelet (PL) structured LiMn_2O_4 .

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