

A Novel Lithium Substitution Induced Tunnel/Spinel Heterostructured Cathode Material for Advanced Sodium-Ion Batteries

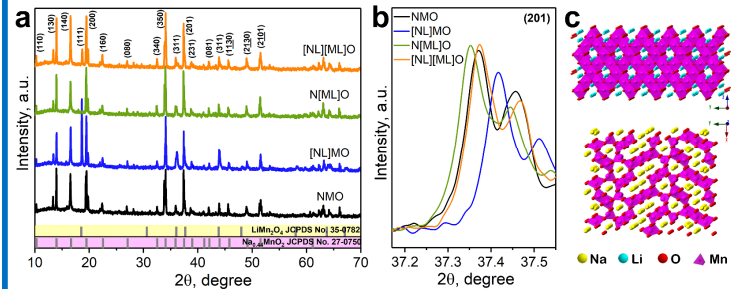
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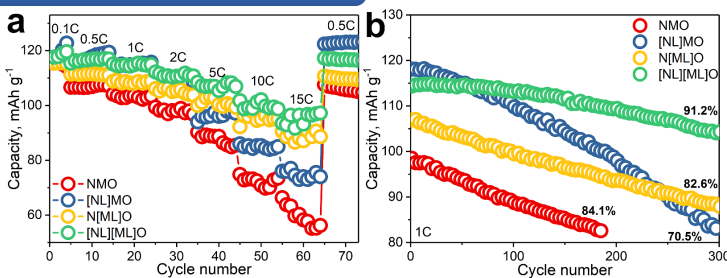
Introduction

The tunnel-type $\text{Na}_{0.44}\text{MnO}_2$ is one of the most promising candidates for advanced sodium-ion batteries, but its commercial application is hindered by poor cycling stability and low high-current capacity due to Jahn–Teller (JT) distortion. Herein, a Li-substituted, tunnel/spinel heterostructured cathode is successfully synthesized for addressing these limitations. In this work, the Li dopant acts as a pillar inhibiting unfavorable multiphase transformation, improving the structural reversibility, and Na storage performance of the cathode. Meanwhile, the tunnel/spinel heterostructure provides 3D Na^+ diffusion channels to effectively enhance the redox reaction kinetics. Therefore, the optimized $[\text{Na}_{0.396}\text{Li}_{0.044}][\text{Mn}_{0.97}\text{Li}_{0.03}]\text{O}_2$ composite delivers an excellent rate performance and cycle performance, demonstrating the potential of the cathode for practical applications.

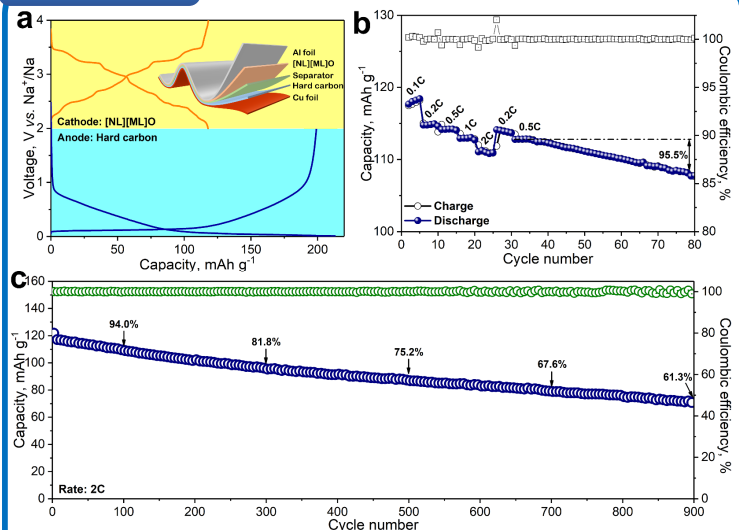
Synthesis and Characterization


Figure 1. Material characterization of the as-synthesized materials.

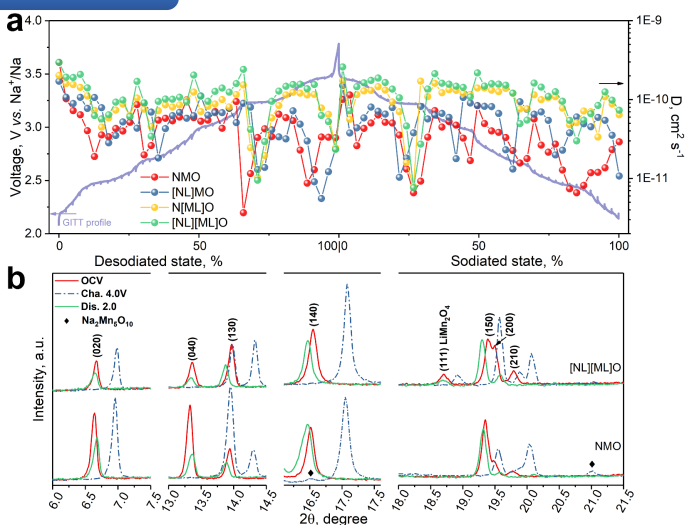
Na half-cell performance


Figure 2. The electrochemical performance of prepared cathodes in coin-type half cells.

Practicality


Figure 3. The electrochemical performances of $[\text{NL}][\text{ML}]\text{O} \parallel$ hard carbon full cells.

Discussion


Figure 4. Electrode process kinetics and structural evolution of the as-prepared cathodes.

Conclusion

- ◆ A tunnel/spinel heterostructured cathode was successfully synthesized for SIBs through Li substitution.
- ◆ The Li dopant acts as a pillar that mitigates structural degeneration and stabilizes the crystal lattice of the host material. And the tunnel/spinel heterostructure provides 3D Na^+ diffusion channels, effectively enhancing the redox reaction kinetics.
- ◆ $[\text{NL}][\text{ML}]\text{O}$ composite delivers a reversible capacity of $119.6 \text{ mA h g}^{-1}$ at 0.1 C with a high Coulombic efficiency of 99.8%. In addition, it demonstrates superior rate capability (15 C) and excellent cycling stability in Na-ion half(1200 cycles, 10C)/full(900 cycles, 2C) battery systems.

References

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- [2] J.-Y. Hwang, J. Kim, T.-Y. Yu, Y.-K. Sun, *Adv. Energy Mater.* 9 (2019) 1803346.
- [3] P. Zheng, J. Su, Y. Wang, W. Zhou, J. Song, Q. Su, N. Reeves-Mclaren, S. Guo, *ChemSusChem* 13 (2020) 1793–1799.