







Solvothermal synthesis of Mn-based MOF materials: Application in high energy density lithium ion battery

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Highlights

- Cobalt free eco-friendly long-run cathode materials for alternate e-vehicles.
- Remarkable cycle stability even after 500 cycles with the capacity retention of 98.3%.
- Complexing agents enhanced the tap density >3.16gcm⁻³.
- High specific capacity of 289mAhg⁻¹.

Abstract

The present work elaborates on the solvothermal synthesis of $\text{Li}[\text{Li}_{0.1}\text{Ni}_{0.3}\text{Mn}_{0.7}]\text{O}_2$ cathode materials derived from metal organic frameworks (LNMO-MOFs) with the novel use of complexing agents, 2-methylimidazole (MI) and 1,10-phenanthroline (PTH). Remarkably, these complexing agents increased the tap density and thereby enhanced the energy density significantly. Additionally, they exhibited low resistance 12.75Ω , compatible particle size distribution (517.46nm) and a specific capacity of 289.03mAhAg^{-1} which are optimal for commercial use. $\text{Li}[\text{Li}_{0.1}\text{Ni}_{0.3}\text{Mn}_{0.7}]\text{O}_2$ surface enabled by 1,10-phenanthroline ensures a unique long cycling stability with a retention of 98.2 % even after 500 cycles. As a result, LNMO-MOFs have been demonstrated to be effective candidates for applications using lithium-ion batteries (LIBs).

Introduction

The growing implementation of energy storage systems and electric vehicles has spontaneously escalated the demand for LIBs. Currently, the need for energy storage has overcome the need for renewable batteries and e-vehicles. For commercial cathode material-production industry, remarkable specific capacity and energy density are crucial in terms of power storage. Layered metal oxide batteries especially the manganese-based lithium-rich ones with high energy density, exceptional specific capacities, low cost and excellent safety have recently drawn drastic interest. Notably, cathode material play significant role in the development of LIBs by directly influencing the energy density, cycle life, cost, safety of the battery etc. Therefore, the objective of this work is to produce cost-effective, eco-friendly, high-energy density, long stability and high specific capacity cathode material for energy storage applications. Hence entropically strong complexing agents such as 2-MI and 1,10-PTH will be incorporated as complexing agents for the controlled preparation of Lithium Nickel Manganese oxide (LNMO) composites.

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Section snippets

Experimental section

For the solvothermal method, two batches of precursor solutions were prepared by dissolving $\text{Mn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (70:30 ratio) in 30ml ethanol followed by addition of 20ml ethanol. The solutions were continuously stirred for 20min at 350rpm in a tank reactor. After complete dissolution, 2ml of complexing agents were added individually and stirred again for about 5min. These metal precursor solutions were transferred into separate autoclaves maintained at 180°C for 12h....

Results and discussion

Potential interactions between metal (Ni, Mn) and complexing agents are shown in Fig. S1. Each complexing agent is expected to establish coordination linkages with transition metal-ions, nickel and manganese since they contain two electron-rich nitrogen sites. This resulted in the formation of their respective metal complexes. Fig. 1a displays the XRD patterns of LNMO impacted by the complexing agents. Majority of peaks are indexed to R3m structure and few peaks between 20° - 30° are indexed to...

Conclusion

LNMO composites were synthesized using the solvothermal method and two different complexing agents, 2-MI and 1,10-PTH. Both the LNMOs had good lamellar structures and were found to be robust. LNMO-1,10-PTH exhibited improved cycle stability and electrochemical performance owed to the inter-twined tightly bound primary particles which led to a highly porous structure, enhanced higher tap density (>3) and varied particle dispersion. In terms of high capacity and rate capabilities, the overall...

CRedit authorship contribution statement

L. Prettenia: Conceptualization, Methodology, Investigation, Writing – original draft, Formal analysis, Visualization. **E. Soundarrajan:** Data curation, Investigation, Formal analysis, Software. **S. Aadheeshwaran:** Data curation, Investigation, Formal analysis, Software. **S. Gnanam:** Data curation, Investigation, Formal analysis, Software. **M. Roselin Ranjitha:** Validation, Writing – review & editing, Formal analysis. **Smagul Zh Karazhanov:** Investigation, Supervision, Validation. **R.A. Kalaivani:**...

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper....

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