

A high-performance and long-cycle-life spinel lithium-ion battery cathode achieved by site-selective doping

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Lithium-ion batteries (LIBs) form an important part of our daily life, powering portable electronic devices, as well as electric and hybrid electric vehicles. However, the limited energy density of current LIBs results in their failure to meet the increasing requirements of rapidly developing technologies. Since the performance limitation in existing LIB technology is the cathode, exploring high-energy-density cathode candidates becomes extremely important. Spinel $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ (LNMO) is considered one of the most promising cathode materials for next-generation high energy-density LIBs, owing to its high operating voltage of 4.7 V vs. Li, low fabrication cost, and high energy density approaching 650 Wh kg⁻¹, which is beyond that of most other LIB cathode materials, such as LiFePO_4 at ~560 Wh kg⁻¹, LiMn_2O_4 at ~480 Wh kg⁻¹, and $\text{LiMn}_{1/3}\text{Ni}_{1/3}\text{Co}_{1/3}\text{O}_2$ at ~510 Wh kg⁻¹. Unfortunately, LNMO suffers from rapid capacity decay and unsatisfactory cycle stability, limiting its practical application and commercialization. Various doping strategies have been widely adopted to enhance the electrochemical stability of LNMO. Although the electrochemical performance of LNMO is enhanced through doping, the mechanism by which the performance is improved remains unclear, with the chemistry- and structure-function relationships for chemically modified LNMOs relatively unknown.

In this work, we not only demonstrate a site-selective doping strategy for an easily-prepared high-performance LNMO cathode through Mg doping, but also comprehensively reveal the underlying enhancement mechanisms using a series of in operando and ex situ characterization techniques. Mg dopants, selectively residing at 8a and 16c sites of the Fd-3m structure, change the way how LNMO responds to the lithium intercalation and de-intercalation during charge-discharge processes. Meanwhile, the addition of Mg ions at such sites significantly prohibits the partially-irreversible two-phase behavior of LNMO, mitigates against the dissolution of transition metals, thus preventing the formation of the undesirable rock-salt phase and reducing the Jahn-Teller distortion and voltage polarization, consequently offering the extraordinary structure stability to LNMO. Consequently, the modified LNMO exhibits excellent extended-long-term electrochemical performance, retaining ~ 86 % and ~ 87 % of initial capacity after 1500 cycles at 1 C and 2200 cycles at 10 C, respectively in half cell configuration, which is reported for the first time and demonstrates their great commercial potential. Such excellent cycle and rate performance are also reflected in a prototype full-battery with a novel TiNb_2O_7 counter electrode. This work provides a new strategy for the chemical modification of electrode materials that may be applied more generally in battery researches, whereby dopants may be used strategically to address specific electrode issues.

Speakers Gender

Male

Level of Expertise

Student

Do you wish to take part in the poster slam

Yes

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