A scalable approach to synthesize cobalt-free LNMO cathode materials for high energy density lithium ion batteries

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ABSTRACT

The rapid growing demand for electric vehicles (EVs) has highlighted the need for high-energy-density batteries. The cathode material plays a vital role in achieving this goal[1]. As the limitations of cathode capacity improvement are approached, the pursuit for high-voltage materials becomes a viable option[2]. Lithium nickel manganese oxide LNMO ($LiNi_{0.5}Mn_{1.5}O_4$) stands out due to its remarkable attributes including a substantial reversible capacity, excellent thermal stability, cost-effectiveness, environmental friendliness, and a high energy density[3]. Moreover, its cobalt-free composition aligns with sustainability objectives. Despite extensive research, the large-scale production and deployment of LNMO remain formidable challenges.

In this study an industrially applicable co-precipitation method-based scalable synthesis strategy is introduced. The LNMO spheres with controlled various sizes and morphologies were successfully fabricated by adjusting the pH environment during synthesis carefully (**Fig. 1A**). Scanning electron microscopy (SEM) revealed the presence of uniformly spherical particles with dimensions of *appr.* 6, 9, and 14 μm and unique morphological characteristics (**Fig. 1A**d-f). Synchrotron X-ray diffraction (SXRD) of LNMO samples revealed a deviation from the optimal Ni-to-Mn ratio of 1:3 (**Fig. 1B**)[4,5]. This variation results from the pH-dependent metal ion precipitation dynamics during synthesis, introducing a fascinating dimension to LNMO material fabrication. As potential cathode materials for lithium-ion batteries, these samples underwent a thorough electrochemical evaluation as part of our research. Among the synthesized LNMO samples, LNMO-9 demonstrated the most promising specific capacity, surpassing approximately 138 mAh/g and working under high voltage of 4.75 V (**Fig. 1C**). Furthermore, the characterizations of the materials were also thoroughly investigated by a variety of advanced characterization techniques, including Raman spectroscopy, X-ray photoelectron spectroscopy (XPS), and transmission electron microscopy (TEM).

This research reveals a strategy for synthesizing LNMO materials with certain regulated features and presents a successful strategy for industrial manufacturing as well. Particularly in the context of the EV industry, this research acquires critical significance in answering the growing demand for high-performance, sustainable energy storage systems.

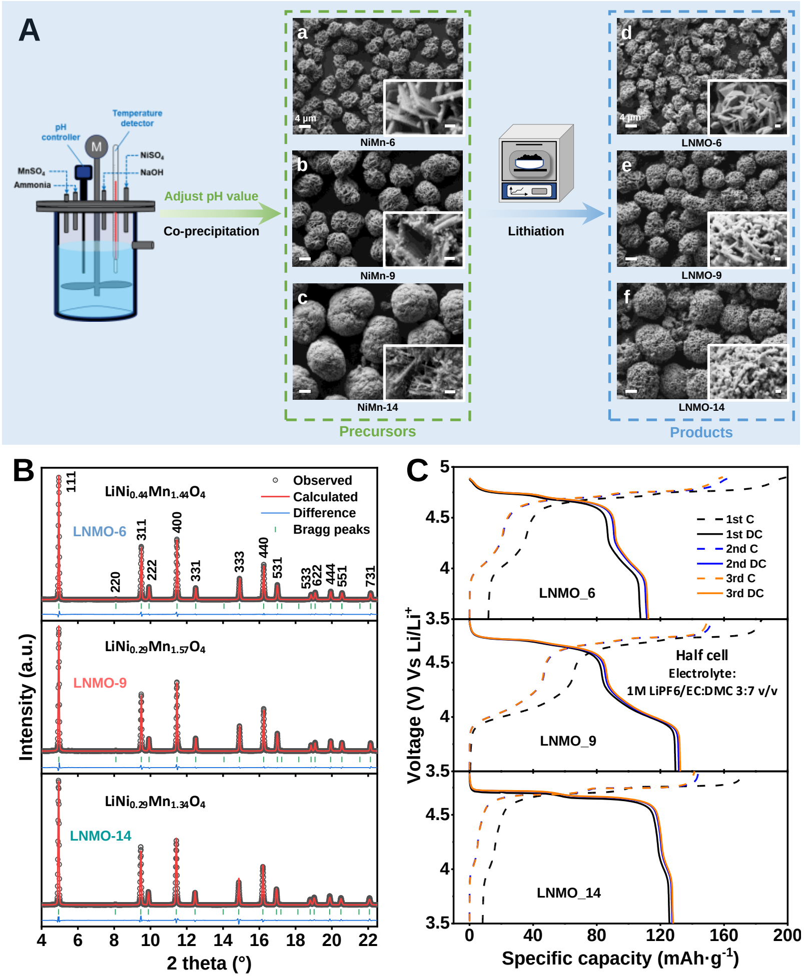

Fig. 1 (A) Illustration of the synthesis process of LNMO-*x* samples with SEM images of (a–c) NiMn-*x* precursors and (d–f) LNMO-*x* products. (B) SXRD patterns with Rietveld refinements and (C) charge/discharge profile of LNMO-*x* samples.

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