

# Medium-frequency induction sintering of lithium nickel cobalt manganese oxide cathode materials for lithium ion batteries



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## ABSTRACT

This study examines the electrochemical performance of  $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$  (LNCM) cathode, prepared by a medium-frequency induction sintering technique. The induction heating method is capable of preparing highly-crystalline LNCM powders. The carbon coating onto LNCM powders appears as a crucial factor in facilitating the specific capacity, rate capability, and cyclic stability. The discharge capacities of C-coated LNCM cathode can reach to 184.6 and 202.3  $\text{mAh g}^{-1}$  at 25 and 55 °C, respectively. This enhanced performance can be ascribed to fast  $\text{Li}^+$  diffusion in the solid solution and electron jumping across the LNCM cathode, with the aid of carbon layer over the LNCM cathode.

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## 1. Introduction

Lithium-ion batteries have received considerable attentions as promising power sources for portable electronic devices and electric vehicles [1,2]. Both energy and power densities of Li-ion battery basically depend on the electrochemical performance of electrode materials. So far, layered  $\text{LiCoO}_2$  serves as a reliable cathode material for commercial Li-ion batteries. However,  $\text{LiCoO}_2$  cathode materials face some critical challenges such as low thermal stability, high toxicity and high cost. The drawbacks direct us to shift research efforts toward the study for alternative cathodes. One strategy is to replace Co usage partially or completely with other transition metals, e.g., Ni or Mn [3]. Recently, one multi-elemental  $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$  (LNCM) cathode has gradually shown commercial feasibility owing to its high specific capacity, superior thermal stability, and especially lower cobalt content making it more effective and less toxic [4,5]. Theoretically, LNCM cathode materials behave as  $\alpha\text{-NaFeO}_2$  crystal structure with a space group of rhombohedral  $R\bar{3}m$  symmetry [6]. The LNCM cathode undergoes a very small volume change of ~3% during  $\text{Li}^+$  insertion/extraction process [7]. In LNCM lattice,  $\text{Ni}^{2+/3+/4+}$  and  $\text{Co}^{3+/4+}$  redox couples display the electrochemical activity, whereas inactive  $\text{Mn}^{4+}$  favors the electrode stability, leading to an improved thermal stability [3,4]. However, most of pioneering studies only emphasized the superior performance of LNCM cathodes at ambient temperature.

Actually, high-temperature operation (usually > 50 °C) delivers a clue in determining the performance, stability and safety of cathodes for various applications of Li-ion batteries, such as electric vehicles and drilling tools [8]. Raising the battery operation temperature is prone to activate electrode materials with higher electronic, ionic, and interfacial conductivities, while the decomposition of materials and side reactions would take place at electrode/electrolyte interface [9]. It is generally recognized that the LNCM cathode still suffers from unsatisfied cycling stability, resulting from its low electronic conductivity, high reactivity between delithiated cathode and electrolyte, and serious dissolution of transition metal ions and electrolyte [10]. Thus, the high-temperature operation of LNCM cathodes becomes more crucial, especially for cycling stability and safety issues. To avoid poor cyclic performance, several approaches have been devoted to synthesize C-coated LNCM cathode materials using different forms of carbon such as graphitized carbon [5, 11], amorphous carbon [12], carbon nanotubes [13] and graphene sheets [6]. The carbon coating improves the electronic conductivity and protects the LNCM cathodes from the electrolyte corrosion. However, there are few reports focusing on the high-temperature cyclic performance of C-coated LNCM cathodes.

Accordingly, this work aims at the influence of operating temperature on the electrochemical performance of C-coated LNCM cathode materials at 25 and 55 °C. Different synthesis methods of LNCM cathode materials have been reported previously [14–19]. This present study adopts one facile synthesis of LNCM cathode using medium-frequency induction heating method. The induction heating is a feasible route for the sintering of advanced materials such as  $\text{LiFePO}_4$  [20],  $\text{Fe}_3\text{O}_4$  [21], and TiC/NiAl [22]. The induction heating in the synthesis of cathode

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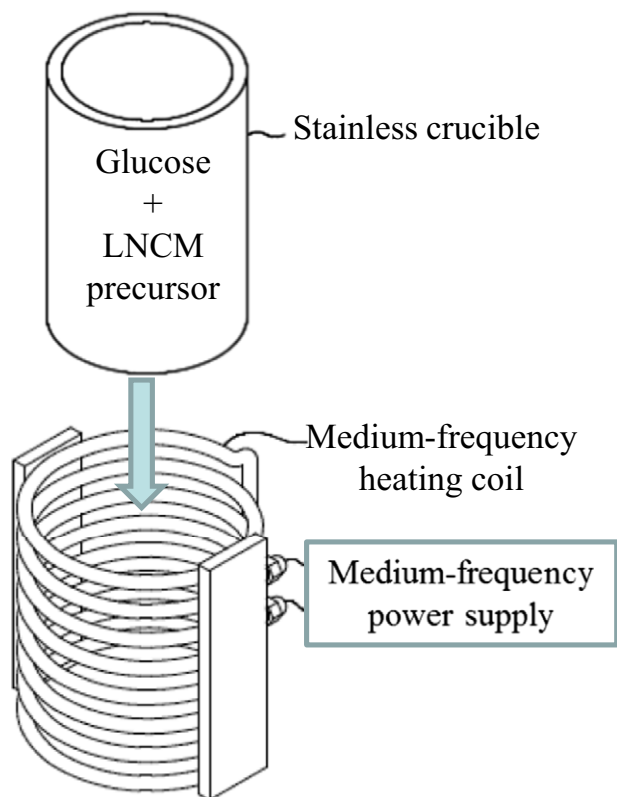


Fig. 1. Schematic illustrating the synthesis of LNCM powders by using medium-frequency induction heating method.

materials can significantly reduce the energy consumption and the overall reaction time with direct impact on the final price [23]. In the induction heating, electromagnetic energy is converted into heat energy which is concentrated within a thin layer termed the penetration depth whose thickness [24]. Pioneering studies have reported that a novel rapid synthesis method for C/LiFePO<sub>4</sub> cathode material that combines

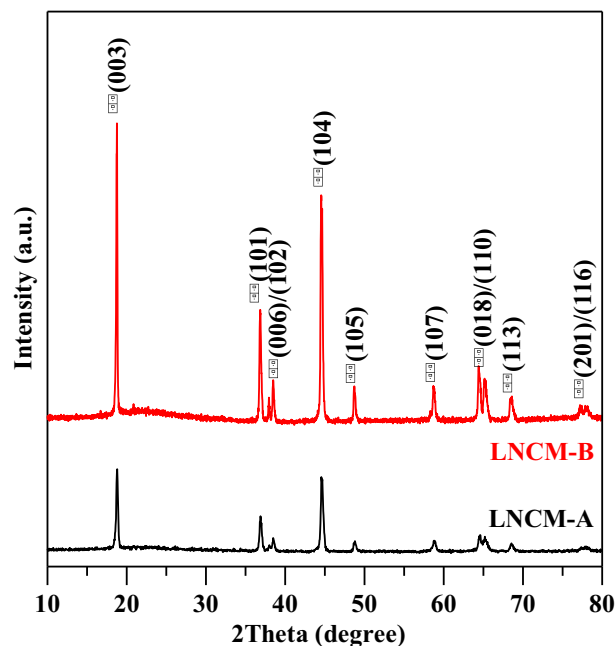


Fig. 3. Typical XRD patterns of (a) LNCM-A and (b) LNCM-B powders.

a carbothermal reduction with a high-frequency induction heating method to reduce its process costs [23,25]. However, medium-frequency induction heating method is seldom reported to prepare LNCM cathode materials. In comparison with other traditional sintering method with a long period (e.g., ~10–15 h), the induction sintering method can be regarded as a fast and economical method for the LNCM preparation. In this study, a conductive crucible made of stainless steel provides rapid heating ( $\sim 200\text{ }^{\circ}\text{C min}^{-1}$ ) by high frequency induction, and the LNCM precursor is heated indirectly by the heat-radiating crucible. The induction heating method is one solid-state sintering process for forming highly-crystalline LNCM powders and carbon coating despite the rapid synthesis only takes 3 h. Herein glucose serves as carbon precursor for depositing nanosized carbon layer onto the LNCM

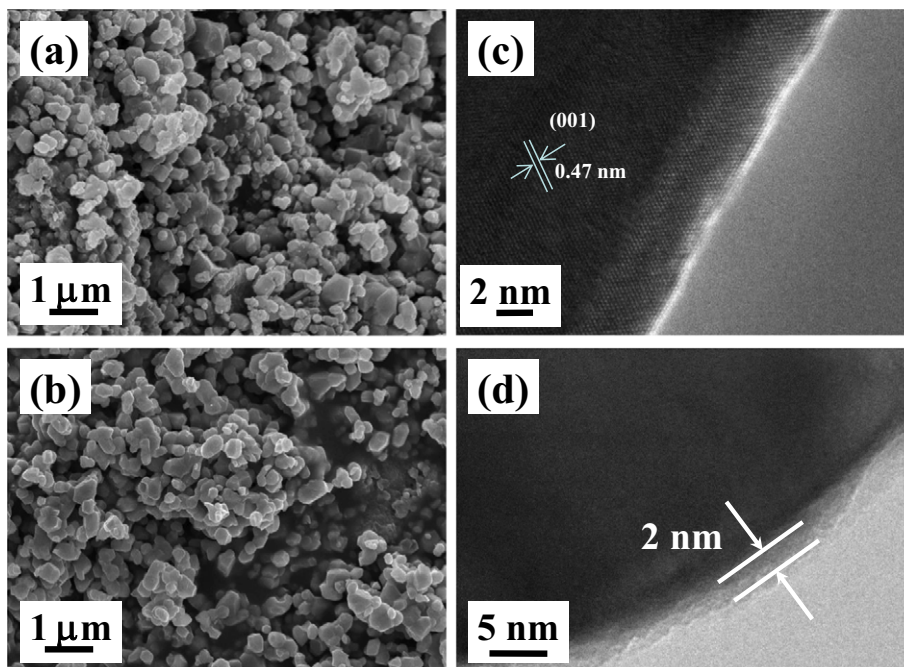


Fig. 2. FE-SEM photographs of (a) LNCM-A and (b) LNCM-B powders. HR-TEM micrographs of (c) LNCM-A and (d) LNCM-B powders.

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