



Effects of Li_2SiO_3 coating on the performance of $\text{LiNi}_{0.5}\text{Co}_{0.2}\text{Mn}_{0.3}\text{O}_2$ cathode material for lithium ion batteries



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ABSTRACT

Layered $\text{LiNi}_{0.5}\text{Co}_{0.2}\text{Mn}_{0.3}\text{O}_2$ (NCM523) material has been functionally coated with a uniform and thin layer of Li_2SiO_3 via a two-step method. Owing to its high lithium ion conduction and excellent structural stability against electrolyte decomposition, Li_2SiO_3 could greatly improve the Li^+ ion diffusion rate and ameliorate the electrochemical capability of the layered oxide materials. Electrochemical tests illustrate that Li_2SiO_3 used as a Li^+ -ion conductor greatly improves electrochemical performance of the NCM523 cathode at high current density under high cutoff voltage. Particularly, the Li_2SiO_3 -modified sample delivers an initial capacity of 140.0 mAh g^{-1} and remains 134.1 mAh g^{-1} even at a high current density of 10 C after 100 cycles, while the capacity of the pristine decreased sharply to 81.5 mAh g^{-1} . The capacity retention of Li_2SiO_3 -modified NCM523 is 96.1%, while only 55.3% for the bare sample. This result demonstrates an efficient method for the Li_2SiO_3 -modified NCM523 cathode with enhanced electrochemical performance, which has a certain reference for other cathode materials of Li-ion batteries.

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1. Introductions

Nickel-rich layered oxide materials $\text{LiNi}_x\text{Mn}_y\text{Co}_{1-x-y}\text{O}_2$ (NCM) have attracted more and more attention as cathode materials for lithium-ion batteries (LIBs), due to their lower cost, higher specific capacity and better thermal stability than those of LiCoO_2 [1,2]. Layered cathode materials $\text{LiNi}_{0.5}\text{Co}_{0.2}\text{Mn}_{0.3}\text{O}_2$, as one of the representatives, have been applied in many commercial cells [3]. However, there are still some drawbacks to restrict its sustainable and high-power applications, such as poor rate capability, low cycle stability at high voltage or at high current density [4,5]. Specifically, when the cells are charged to 4.5 V, the high valent Ni^{4+} ions are highly active, which could cause the oxidative degradation of the electrolyte, thus leading to the poor cycle performance [6].

To overcome these problems, much efforts have been made to ameliorate the electrochemical properties of the cathodes, such as surface modification and ion substitution into the lattice [7,8]. Particularly, coating materials for surface modification, such as AlF_3 [9], MgO [10], Al_2O_3 [11], Li_3PO_4 [12], SiO_2 [13], ZrO_2 [14], Li_2VO_3 [15], AlPO_4 [16], TiO_2 [17] and so on [18–22], has also been employed in many cathodes. Unfortunately, the majority of shell

cladding materials are usually Li-ion insulator, which is not beneficial for facile Li-ion transportation during cycling at the interfacial of the electrode [23].

Therefore, an ideal coating material should have some unique advantages compared with the abovementioned coating materials, which could stabilize the structure of cathode materials and possess high Li^+ -ion conduction [24]. And Li_2SiO_3 is such a kind of ideal coating material. Due to its chemically inert substance, Li_2SiO_3 can ensure high structural stability in organic electrolyte, which can protect the bulk materials from the erosion of HF as a protective layer [25–27]. At the same time, Li_2SiO_3 can provide a three-dimensional network of channels beneficial for Li^+ -ion migration which can effectively increase Li^+ -ion conduction, and thus ameliorate the rate capability of the cathode materials. Zhao et al. [27] reported that Li_2SiO_3 -coated $\text{Li}_{1.13}\text{Ni}_{0.30}\text{Mn}_{0.57}\text{O}_2$ exhibited enhanced electrochemical performances and low interfacial polarization, due to a uniformly conductive Li_2SiO_3 coating layer. Atsushi et al. [28] reported that the coating layer of Li_2SiO_3 was more effective than the SiO_2 in enhancing rate property, indicating that lithium-ion conductivity of the coating materials is important for high-rate performance.

Based on the above advantages, a unique and complete Li_2SiO_3 coating layer is expected to significantly ameliorate the electrochemical properties and reduce the polarization of layered cathode materials. In this work, we used Li_2SiO_3 as a coating material and

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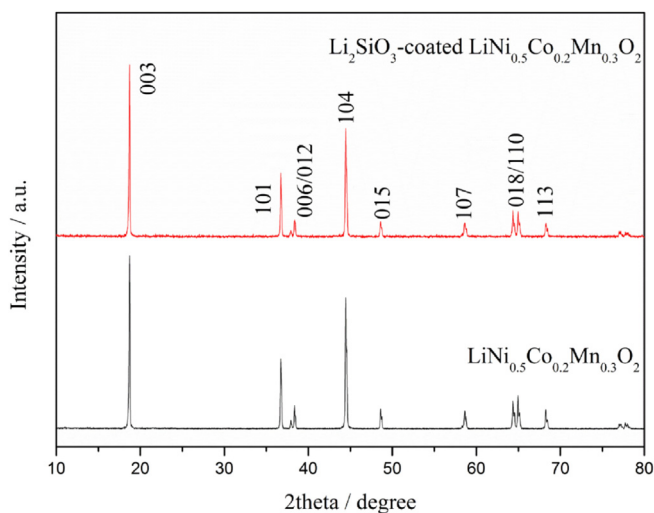


Fig. 1. XRD images of the bare and Li_2SiO_3 -coated $\text{LiNi}_{0.5}\text{Co}_{0.2}\text{Mn}_{0.3}\text{O}_2$ materials.

employed a two-step synthesis method to successfully make a uniform and consistent layer of Li_2SiO_3 on the surface of NCM523. In addition, the structure and morphology of the samples were systematically analysed to explore the impact of the Li_2SiO_3 on the modified NCM523 material. As a result, the modified sample showed improved rate and cycling performance when used as the cathode for LIBs.

2. Experimental methods

2.1. Preparation of Li_2SiO_3 -coated $\text{LiNi}_{0.5}\text{Co}_{0.2}\text{Mn}_{0.3}\text{O}_2$

First, the $\text{LiNi}_{0.5}\text{Co}_{0.2}\text{Mn}_{0.3}\text{O}_2$ cathode material was coated with SiO_2 through a hydrolysis process. For this purpose (Calculated by 1.0 wt% Li_2SiO_3 amount), 10 g of $\text{LiNi}_{0.5}\text{Co}_{0.2}\text{Mn}_{0.3}\text{O}_2$ was dispersed

into 20 mL of ethanol and deionized water solution (9:1, in volume), followed by addition of ammonia solution and $\text{Si}(\text{OC}_2\text{H}_5)_4$ (Xilong Chemical, AR) (TEOS, 0.26 mL) diluted into 10 mL ethanol solution respectively. The resulting precipitate was collected and washed with ethanol to remove residual TEOS and then dried under vacuum at 60 °C. After that, the precipitate and lithium acetate (Aladdin, AR 99.0%) (0.15 g) was dissolved in 30 mL ethanol solution and stirred vigorously at 80 °C until the solvent completely evaporated. And then the solid powder was dried at 120 °C for one night. Finally, the obtained product was scraped off and calcined at 700 °C for 5 h with a heating rate of 2.5 °C min^{-1} to obtain Li_2SiO_3 -coated $\text{LiNi}_{0.5}\text{Co}_{0.2}\text{Mn}_{0.3}\text{O}_2$ sample.

2.2. Material characterizations

The crystal structures of all the samples were characterized by X-ray diffraction (XRD, Rigaku D/maxb) equipped with $\text{Cu K}\alpha$ radiation under test in the range of 10°–80° at a scan speed of 10° min^{-1} . Particle morphologies of the as-prepared powders were observed by scanning electron microscope (SEM, JEOL JSM-6360LV) and transmission electron microscopy (TEM, FEI TECNAI G2 F20). The distributions and oxidation states of related elements throughout the powder were obtained using an energy dispersive spectrometer method (EDS) and X-ray photoelectron Spectrometer method (XPS, Thermo Fisher-VG Scientific ESCALAB 250Xi).

2.3. Electrochemical measurements

To fabricate the positive electrodes, the process was referred to our previous report [29]. After that, the positive electrodes were dried for 12 h at 120 °C in vacuum drying oven. Prior to use, the positive electrodes were electrochemically measured under the test using a 2025-type coin cells with Li film as the corresponding counter electrodes. And the electrolyte solution consisted of 1 M LiPF_6 in EC-DEC-EMC (1:1:1, v%) and the separator was Celgard 2400. Galvanostatic charge-discharge cycling tests were measured

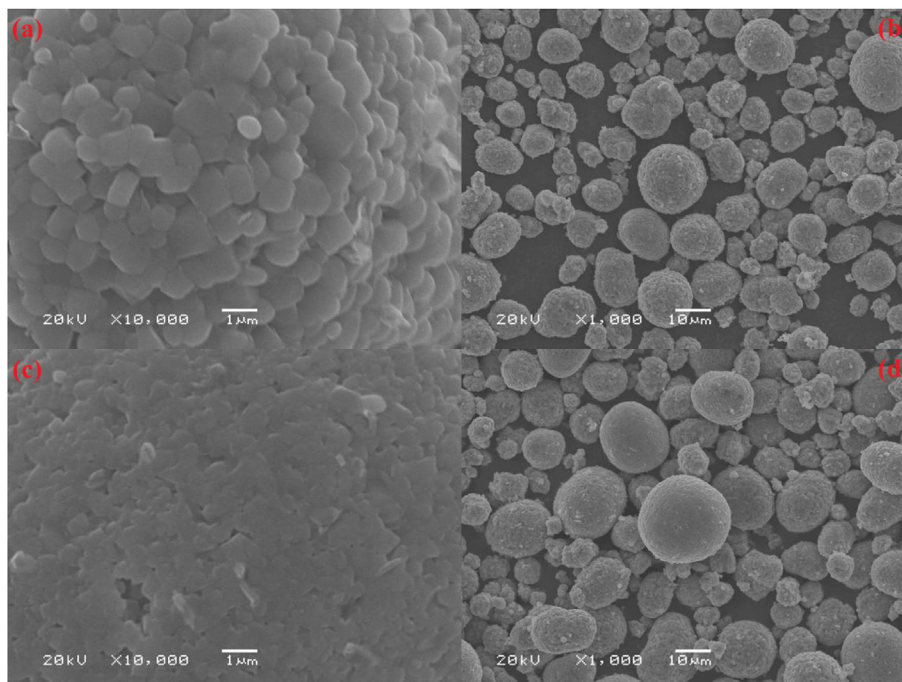


Fig. 2. SEM images of (a, b) the bare and (c, d) Li_2SiO_3 -coated $\text{LiNi}_{0.5}\text{Co}_{0.2}\text{Mn}_{0.3}\text{O}_2$ materials.

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