Surface Coating of NCM-811 Cathode Materials with $g$-$C_3N_4$ for Enhanced Electrochemical Performance

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ABSTRACT: Li(Ni$_{0.8}$Co$_{0.1}$Mn$_{0.1}$)O$_2$ (NCM-811) cathode materials have been commercialized recently, aiming to increase the specific capacity and specific energy of the lithium-ion battery for practical applications in electric vehicles. The surface coating has been proved to be an effective approach for the stabilization of NCM-based cathodes, which could reduce the structural instability and prevent surface reactions between the cathode materials and electrolytes. Herein, we demonstrate the facile synthesis of graphitic carbon nitride ($g$-$C_3N_4$)-coated NCM cathodes with both the sonication-assisted liquid exfoliation method ($g$-$C_3N_4$NS@NCM-811) and chemical vapor-assisted coating method ($g$-$C_3N_4$@NCM-811). It is discovered that coating with a thin $g$-$C_3N_4$ layer could reduce the impedance of the NCM-811 cathode material, as well as increase the cycle stability of the cathode material, leading to increased capacity retention from 130 mA h/g (for the pristine sample) to 140 mA h/g after 225 cycles. While the coating of thick $g$-$C_3N_4$ nanosheets could hinder the lithium intercalation, resulting in significant loss of specific capacity. This study paves the way toward practical applications of the $g$-$C_3N_4$-coated NCM-811 cathode materials.

1. INTRODUCTION

The recent rapid development of electric vehicles requires tremendous efforts to improve the specific energy of lithium battery systems to solve the “mileage anxiety.” Among all the commercially available lithium-ion battery cathode materials, Ni-rich-layered oxides (LiNi$_x$Co$_y$Mn$_z$O$_2$, denoted as NCM, where $x + y + z = 1$) exhibit the highest specific energy density,$^{1-5}$ which has been considered as the potential next-generation cathode materials for the complete electrification of transportation. In particular, Li(Ni$_{0.8}$Co$_{0.1}$Mn$_{0.1}$)O$_2$ (denoted as NCM-811) has been commercialized recently, which could significantly minimize the use of expensive Co resources while also ensuring considerable stability of the battery system.$^6-9$ Meanwhile, it is well known that during the de-lithiation process, the transition-metal ions could dissolve into the electrolyte due to the surface side reactions (e.g., with the residual water or HF in the electrolyte), leading to structural instability and capacity fade.$^9-11$

The surface coating has been proved to be an effective avenue for the stabilization of NCM-based cathode materi-
raw materials, which hinder the practical commercial applications.

Graphitic carbon nitride (g-C₃N₄) has been shown to exhibit excellent chemical resistance and a moderate band gap (~2.7 eV)²⁻⁵⁰, which has been widely explored for applications in photocatalysis,²⁴⁻²⁵ sodium-ion battery anode,²⁶ lithium-ion battery anode,²⁷ separator for lithium-sulfur battery,²⁸ and so forth. In particular, the excellent chemical resistance and moderate band gap of g-C₃N₄ make it a good potential coating material for the NCM cathode, which is stable in the electrolyte during cycling, while also ensuring proper electronic and ionic transport across the C₃N₄/NCM interface. Moreover, g-C₃N₄ has a similar layered structure to graphite, where the layers are bound relatively weak van der Waals forces. For g-C₃N₄, the lattice spacing between layers is about 3.26 Å,²⁹⁻³¹ which can act as a channel for Li⁺ diffusion.

Herein, in this study, we designed a surface-coating routine of the NCM-811 cathode materials with graphitic carbon nitride (g-C₃N₄) via both sonication-assisted liquid exfoliation (g-C₃N₄NSs@NCM-811) and chemical vapor-assisted coating methods (g-C₃N₄@NCM-811). It is discovered that coating with a thin g-C₃N₄ layer (thin g-C₃N₄@NCM-811, ~20 nm) could reduce the impedance of the NCM-811 cathode material, as well as increase the cycle stability of the cathode material, leading to the increase of specific capacity from 130 mA h/g (for the pristine sample) to 140 mA h/g after 225 cycles. While the thick coating of g-C₃N₄ (thick g-C₃N₄@NCM-811, ~70 nm) could lead to a large decrease of specific capacity (decreases to ~115 mA h/g after 225 cycles). Furthermore, the g-C₃N₄ nanosheet coating (g-C₃N₄NSs@NCM-811) by simple mechanical ball milling forms a much thicker (~100 nm) and inhomogeneous coating, which could hinder the intercalation of Li⁺, resulting in almost no energy storage capacity.

2. EXPERIMENTAL SECTION

2.1. Materials Synthesis. The schematic illustration of the synthesis process for the g-C₃N₄-coated NCM-811 cathode materials is shown in Figure 1. Two types of cathode materials are designed, namely, the g-C₃N₄ nanosheet-coated NCM-811 (denoted as g-C₃N₄NSs@NCM-811) and g-C₃N₄ surface-coated NCM-811 (denoted as g-C₃N₄@NCM-811). To synthesize the g-C₃N₄NSs@NCM-811 (Figure 1a), melamine (99%, Shanghai Macklin Biochemical Co., Ltd.) was first sintered and decomposed at 550 °C to obtain g-C₃N₄. Then, 400 mg yellowish g-C₃N₄ particle was dispersed in 200 ml deionized water. The sonication-assisted liquid exfoliation method was further employed, where the water/g-C₃N₄ mixture was treated with sonication for 75 min to form a g-C₃N₄ nanosheet suspension. Then, the suspension was purified via centrifugation at a speed of 5000 rpm for 5 min. Consequently, 400 mg NCM-811 (purchased from Ningbo Rongbay New Energy Technology, Co. Ltd., China) was added, stirred vigorously with a magnetic stirrer, filtered, and dried at 80 °C for 12 h. Meanwhile, to obtain the g-C₃N₄@NCM-811, a chemical vapor-assisted synthesis method is used (Figure 1b). Different amounts of melamine were added to control the thickness of coating layers, where 1 g/2 g melamine powder and 5 g NCM-811 powder were put separately in a closed crucible and heated to 350 °C for 2 h to ensure the full vaporization of melamine. Then, they were further heated to 550 °C for 4 h to decompose the surface melamine to form g-C₃N₄. The thin g-C₃N₄@NCM-811/thick g-C₃N₄@NCM-811 can be obtained after furnace cooling for the samples with 1 g/2 g melamine powder.

2.2. Structural Characterization. Structural characterizations were performed to investigate the phase, morphology, and elemental distributions of the cathode materials. X-ray diffraction (XRD) measurements were employed with Cu-Kα (MiniFlex 600 Diffractometer, Rigaku, Japan, λ = 1.54059 Å) to unveil the crystal structure of the coated and uncoated NCM-811, with a scan rate of 10°/min in the range of 5°–90°. The surface morphology was further characterized by field-emission scanning electron microscopy (SEM, Hitachi S-4800, 5 kV) and high-resolution transmission electron microscopy (HRTEM, Hitachi HT-7700, 120 kV).

2.3. Battery Assembly. The cathode was obtained with 80 wt % of the as-prepared NCM-811 materials, 10 wt % Ketjen Black (Lion Co., Japan), and 10 wt % polyvinylidene fluoride (PVDF, Arkema, France). They were ground for 10 min in an agate mortar to ensure full mixing before dissolving in a NMP (99.5%, Shanghai Aladdin Bio-Chem Technology Co., Ltd., China) solvent and consequently stirred vigorously for 5 h to form a homogenous slurry. It was further cast on an Al foil and dried at 80 °C for 12 h in vacuum before pocking into round cathode disks. They were then calendared under 20 MPa and dried in vacuum at 80 °C for another 4 h to further remove the residual NMP solvent and water. The CR2025 standard coin cells were further assembled, with a Li metal anode (China Energy Lithium Co., Ltd., Tianjin, China) as the counter electrode and 1.0 M LiPF₆ in EC/DMC = 3:7 vol % with 5.0% FEC additives (Suzhou Duoduo Chemical Tech. Co., Ltd., China) as the electrolyte and separator (Celgard Co. Ltd., USA).

2.4. Electrochemical Testing. The electrochemical performances were tested using a Land battery testing station (Wuhan LAND Electric Co. Ltd., Wuhan, China) under a voltage range of 2.6–4.2 V. The first cycle was tested at 0.1 C, followed by 1 C for the consequent cycles (1 C = 205 mA/g).

Figure 1. Schematic illustrations of the synthesis process for (a) g-C₃N₄NSs@NCM-811 and (b) g-C₃N₄@NCM-811.
3. RESULTS AND DISCUSSION

XRD measurements are performed to unveil the phase of the material. As shown in Figure 2a, the XRD pattern for the pristine NCM-811 matches well with a pure $\alpha$-NaFeO$_2$-type crystal structure (space group $R\overline{3}m$, JCPDS #09-0063) with no obvious secondary phase peaks. The corresponding crystal surfaces for the major peaks have been indexed in the figure. The splitting of the two peaks, namely, (006)/(012) and (018)/(110), can be attributed to the hexagonal ordering of the layered structure for NCM-811. Meanwhile, it is also widely accepted that the intensity ratio of the (003)/(104) peaks is determined by the degree of cation disorder, where the lower intensity ratio leads to a higher degree of Li$^+$/Ni$^{2+}$ cation disorder. The measured intensity ratio of the (003)/(104) peaks for the pristine sample is 1.4, showing a cation-ordered phase. Both the g-C$_3$N$_4$NSs@NCM-811 and g-C$_3$N$_4$@NCM-811 exhibit a similar pattern to the pristine sample, with a slight change in the peak intensities, indicating that the majority of the phase of the material does not change during the coating procedures. The intensity ratio of the (003)/(104) peaks for the g-C$_3$N$_4$NSs@NCM-811 and g-C$_3$N$_4$@NCM-811 decreases to 1.3 and 1.2, respectively. This reveals that the coating procedure, in particular, the heat treatment, could increase the Li$^+$/Ni$^{2+}$ cation disorder. This can be attributed to the oxygen release during the heat treatment. While it is reported that increasing the cation disorder leads to a poorer rate performance and cycle performance due to its hindrance to Li$^+$ transport with disordered Ni$^{2+}$. Because the intensity ratio only changes slightly, the influence of the cation disorder on the cathode performance should be minimal. Notably, no obvious C$_3$N$_4$ peaks can be discovered in both samples. It can be understood that because the g-C$_3$N$_4$ coating layer is very thin it cannot be detected by the XRD technique.

Figure 2. Structural characterizations for the coated/uncoated NCM-811 cathode materials. (a) XRD patterns of the cathode materials. SEM image of (b) pristine NCM-811 particle, (c) g-C$_3$N$_4$NSs@NCM-811, (d) thin g-C$_3$N$_4$@NCM-811, and (e) thick g-C$_3$N$_4$@NCM-811. (f–i) Magnified view of (b–d), respectively. EDS mapping of (j) g-C$_3$N$_4$NSs@NCM-811, (k) thin g-C$_3$N$_4$@NCM-811, and (l) thick g-C$_3$N$_4$@NCM-811 particle, the Ni, Co, Mn, and N elements are shown.
The SEM images for the pristine NCM-811, \( \text{g-C}_3\text{N}_4\text{NSs@NCM-811} \), thin \( \text{g-C}_3\text{N}_4\text{@NCM-811} \), and thick \( \text{g-C}_3\text{N}_4\text{@NCM-811} \) are displayed in Figure 2b–i. It can be observed that the pristine samples are spherical in shape, with a diameter ranging from 10 to 20 \( \mu \text{m} \) (Figure 2b). The large spherical particle is formed by the assembly of small nanosized primary particles. The particle structure is preserved for the coated NCM-811 samples, as shown in Figure 2c–e. Meanwhile, the magnified view of the \( \text{g-C}_3\text{N}_4\text{NSs@NCM-811} \) shows the formation of a thick coating layer on the NCM-811 surface (Figure 2g). Whereas the surface coating layer is not as obvious in the \( \text{g-C}_3\text{N}_4\text{@NCM-811} \) (Figure 2h,i). The EDS mappings for the \( \text{g-C}_3\text{N}_4\text{NSs@NCM-811} \) (Figure 2j), thin \( \text{g-C}_3\text{N}_4\text{@NCM-811} \) (Figure 2k), and thick \( \text{g-C}_3\text{N}_4\text{@NCM-811} \) (Figure 2k) show the distribution of elements on the surface of the coated samples.

Figure 3. TEM image of (a) pristine NCM-811 particle, (b) \( \text{g-C}_3\text{N}_4\text{NS} \), (c) \( \text{g-C}_3\text{N}_4\text{NSs@NCM-811} \), (d) thin \( \text{g-C}_3\text{N}_4\text{@NCM-811} \), and (e) thick \( \text{g-C}_3\text{N}_4\text{@NCM-811} \).

Figure 4. Electrochemical performance for the coated/uncoated NCM-811 cathode materials. (a) \( \frac{dQ}{dV} \) with respect to electric potential \( (V) \). (b) EIS measurements. (c) Charge/discharge curve. (d) Discharge capacity (hollow dots) and coulombic efficiency (filled dots) during cycling at 1 C.

The SEM images for the pristine NCM-811, \( \text{g-C}_3\text{N}_4\text{NSs@NCM-811} \), thin \( \text{g-C}_3\text{N}_4\text{@NCM-811} \), and thick \( \text{g-C}_3\text{N}_4\text{@NCM-811} \) are displayed in Figure 2b–i. It can be observed that the pristine samples are spherical in shape, with a diameter ranging from 10 to 20 \( \mu \text{m} \) (Figure 2b). The large spherical particle is formed by the assembly of small nanosized primary particles. The particle structure is preserved for the coated NCM-811 samples, as shown in Figure 2c–e. Meanwhile, the magnified view of the \( \text{g-C}_3\text{N}_4\text{NSs@NCM-811} \) shows the formation of a thick coating layer on the NCM-811 surface (Figure 2g). Whereas the surface coating layer is not as obvious in the \( \text{g-C}_3\text{N}_4\text{@NCM-811} \) (Figure 2h,i). The EDS mappings for the \( \text{g-C}_3\text{N}_4\text{NSs@NCM-811} \) (Figure 2j), thin \( \text{g-C}_3\text{N}_4\text{@NCM-811} \) (Figure 2k), and thick \( \text{g-C}_3\text{N}_4\text{@NCM-811} \) (Figure 2k) show the distribution of elements on the surface of the coated samples.
(Figure 2l) show that the distribution of the transition metals (Ni, Co, and Mn) and N elements are homogenous in the particle region, while the N signal is brighter near the particle edge for the g-C$_3$N$_4$NSs@NCM-811 sample. This indicates a thicker and inhomogeneous coating of the g-C$_3$N$_4$ nanosheets on the NCM-811 particle surface with pure mechanical mixing.

TEM characterizations are performed for the pristine NCM-811, g-C$_3$N$_4$ nanosheet, g-C$_3$N$_4$NSs@NCM-811, and g-C$_3$N$_4$@NCM-811, as depicted in Figure 3. As shown in Figure 3a, the pristine NCM-811 particle (shown in black contrast) has a well-defined shape interface without an obvious surface coating layer. The surface rupture can be discovered, which can be attributed to the nanosized primary particles. The g-C$_3$N$_4$ nanosheet prepared by the sonication-assisted liquid exfoliation method is presented in Figure 3b, which forms a flake-like irregular shape with a size of ~100 nm. For the g-C$_3$N$_4$NSs@NCM-811, while the overall shape is similar to the pristine sample, a thick coating layer (~100 nm) can be discovered on the black NCM-811 particle (Figure 3c). It can also be observed that flakes similar to the g-C$_3$N$_4$ nanosheet are further attached to the NCM-811 surface beside the direct surface coating. Meanwhile, a thin homogenous coating layer can be discovered for the thin g-C$_3$N$_4$@NCM-811 (Figure 3d), showing that g-C$_3$N$_4$ has been homogeneously coated on the NCM-811 particle. With the increase in the weight ratio of melamine from 20% to 40%, the g-C$_3$N$_4$ coating layer increases from ~20 to ~70 nm. Overall, the SEM and TEM characterizations confirm the successful surface coating of the g-C$_3$N$_4$ on the NCM-811 particle for both g-C$_3$N$_4$NSs@NCM-811 and g-C$_3$N$_4$@NCM-811, whereas the coating layer is much thicker for g-C$_3$N$_4$NSs@NCM-811. This demonstrates that the thickness of the g-C$_3$N$_4$ coating layer can be adjusted by changing the melamine/NCM-811 ratio.

The electrochemical performance is further tested and given in Figure 4. The differential capacity analysis is performed for the three samples (Figure 4a). It can be found that the pristine NCM-811 exhibits multiple peaks between 3.5 to 4.2 V for both charge and discharge cycles, consistent with previous reports. A slight right shift can be discovered for the g-C$_3$N$_4$@NCM-811, which can be attributed to the slightly increased overpotential with surface coating. Whereas no peaks can be discovered for the g-C$_3$N$_4$NSs@NCM-811, indicating that the coating of thick g-C$_3$N$_4$ nanosheets could hinder the intercalation process for the NCM-811 cathodes. The electrochemical impedance spectroscopy (EIS) measurements are further performed, which demonstrates increased charge-transfer resistance ($R_{ct}$) from 412.7 Ω to 498.1 Ω for the g-C$_3$N$_4$NSs@NCM-811 as compared to the pristine sample. While the series resistance $R_s$ also increases from around 2.0 to 14.1 Ω for the g-C$_3$N$_4$NSs@NCM-811 (Figure 4b). This indicates that thick g-C$_3$N$_4$ nanosheet coating would increase both series resistance and charge-transfer resistance, which can be understood as because of bulk g-C$_3$N$_4$ being a semiconductor with a band gap of 2.7 eV. Interestingly, the charge-transfer resistances of the g-C$_3$N$_4$@NCM-811 samples reduce from 412.7 Ω to around 320 Ω. The reduced charge-transfer resistance indicates the prohibition of the surface side reactions with electrolytes, which could improve the surface stability and the electrochemical performance. The charge–discharge profile is presented in Figure 4c. The charge profile for the pristine sample and g-C$_3$N$_4$@NCM-811 almost overlap with each other, with a plateau of ~3.75 V and a linear slope between 3.8 to 4.2 V, corresponding to the multiple peak configurations in the dQ/dV curve. While for the discharge curve, the thin g-C$_3$N$_4$-coated cathode shows a slightly lower capacity under the same voltage above 3.7 V, while a higher capacity below 3.7 V, indicating that the coating could enhance the overall structural stability during the discharge process. As the coating layer of g-C$_3$N$_4$ becomes thicker, the specific capacity of cathode materials decreases. Meanwhile, the g-C$_3$N$_4$NSs@NCM-811 shows almost no plateau and capacity during the charge and discharge. The specific capacity and coulombic efficiency during cycling are further given, under 1C, both pristine NCM-811 and thin g-C$_3$N$_4$-coated NCM-811 exhibit an initial specific discharge capacity of ~170 mA h/g, consistent with a previous report (Figure 4d). While the specific capacity almost coincides for the first 50 cycles, the thin g-C$_3$N$_4$@NCM-811 demonstrates higher capacity retention after 50 cycles. It is shown that after 225 charge/discharge cycles, the discharge capacity decreases to ~130 mA h/g for the pristine NCM-811, whereas the g-C$_3$N$_4$@NCM-811 shows a higher capacity of ~140 mA h/g, proving that the coating could increase the overall cycle stability of the NCM-811 cathodes. The improved stability during cycling can be attributed to two factors: (1) g-C$_3$N$_4$ can act as an artificial CEI, which can prevent the dissolution of transition-metal ions on the electrolyte. (2) g-C$_3$N$_4$ can reduce the charge-transfer resistance because it prohibits the surface reaction between the NCM cathode and electrolyte that could form insulating CEI components such as LiF. Whereas for the g-C$_3$N$_4$NSs@NCM-811, the specific capacity is ~5 mA h/g, indicating that the intercalation process is hindered after the surface coating of thick g-C$_3$N$_4$ nanosheets. It is worthwhile to note that coulombic efficiency during the cycling for both thin and thick g-C$_3$N$_4$ coating increases, which demonstrates that the g-C$_3$N$_4$ coating layer can prevent the side reactions between the NCM-811 cathode and electrolyte.

4. CONCLUSIONS

In conclusion, we have synthesized the g-C$_3$N$_4$-coated NCM-811 cathode materials via the sonication-assisted liquid exfoliation method (g-C$_3$N$_4$NSs@NCM-811) and chemical vapor-assisted coating method (g-C$_3$N$_4$@NCM-811). It is discovered that the surface coating of a thin g-C$_3$N$_4$ layer (thin g-C$_3$N$_4$@NCM-811) could reduce the impedance of the NCM-811 cathode materials, as well as increase the cycle stability of the cathodes, leading to increased capacity retention from 130 mA h/g for the pristine sample to 140 mA h/g after 225 cycles. Meanwhile, for the g-C$_3$N$_4$NSs@NCM-811, the impedance increases while the specific capacity is ~5 mA h/g, showing that the lithium intercalation is severely hindered by the thick coating layer. We hope this study could spur further interest in the research and practical applications of the g-C$_3$N$_4$ surface-coated NCM-811 cathodes.

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Z. H. would like to acknowledge a start-up grant from Zhejiang University.

ACKNOWLEDGMENTS

Z. H. would like to acknowledge a start-up grant from Zhejiang University.

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