北京理工大学研究生学籍异动申请表

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北京理工大学研究生课程成绩单

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0900037	计算材料学与材料设计	3	48	91
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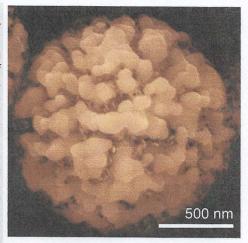




Fast Capacitive Energy Storage and Long Cycle Life in a Deintercalation-Intercalation Cathode Material

Lecai Wang^a, Li Li^{a,b}, Hanyong Wang^a, Jingbo Yang^a, Yitian Ma^a, Jiawei Wu a, Feng Wu^{a,b}, Renjie Chen^{a,b}*

Abstract: Ni-rich Li-ion cathode materials promise high energy density. but are limited in power density and cycle life, resulting from their poor dynamic characteristics and quick degradation. On the other hand, capacitor electrode materials promise high power density and long cycle life but limited capacities. A joint energy storage mechanism of these two kinds is performed in the material-compositional level in this paper. A valence coupling between carbon π-electrons and O2- is identified in the as-prepared composite material, using a tracking X-ray photoelectron spectroscopy strategy. Besides delivering capacity simultaneously from its LiNi0.8Co0.1Mn0.1O2 and capacitive carbon components with impressive amount and speed, this material shows robust cycling stability by preventing oxygen emission and phase transformation via the discovered valence coupling effect. Structural evolution of the composite showed a more flattened path compared to that of the pure LiNi0.8Co0.1Mn0.1O2, revealed by the in-situ X-ray diffraction strategy. Without obvious phase transformation and losing active contents in this composite material, long cycling can be achieved.



Reviewer #1: The authors proposed a way to integrate capacitive carbon material with NMC811 material for high cycling stability and high power performance. Although similar ideas may have been demonstrated using other materials, it is impressive to see the excellent performance demonstrated in this paper. I am ok to recommend the publication of the paper but would like the authors to help me understand the work better. My questions are as below.

- 1) Will the NMC811 get reduced during the carbonization? NMC811 is also very sensitive to H20. Will its structure be damaged during the annealing with H20 and CO2 gas?
- 2) The capacitive carbon content seems low according to the TGA. I am curious how much contribution from it?
- 3) What is the electrode loading for the high rate tests?
- 4) What do the bulk and hierarchical NMC811 look like under SEM and TEM?

Reviewer #2:This manuscript investigated the composite between 811 and carbon. This composite showed better rate capability and capacity retention compared with bare 811. From XPS and XRD, it appears that the presence of carbon altered the phase transformation of 811 at high voltages, and thus inhibiting oxygen loss. It is also funny to see these authors are making analogy with Iron Man and Flash. I found this paper interesting overall, and would like to recommend the paper to Small. I have one question, though, what is the difference between carbon reported in this work and other types of carbon reported in literature. Clearly, not all carbon bring similar effects. Can the authors elaborate a bit more on this?

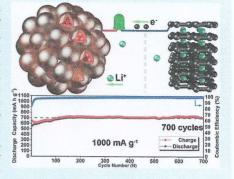
Stable Conversion Mn₃O₄ Li-Ion Battery Anode Material with Integrated Hierarchical and Core-Shell Structure

Lecai Wang,[†] Li Li,^{†,‡} Hanyong Wang,[†] Jingbo Yang,[†] Feng Wu,^{†,‡} and Renjie Chen*,^{†,‡}

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Supporting Information

ABSTRACT: Anodes composed of Mn₃O₄ deliver a much higher specific capacity in Li-ion batteries (LIBs) than that of commercial graphite but suffer from poor cycling stability, a poor rate characteristic, and a high overpotential stemming from volumetric changes during cycling, low electroconductibility, and insufficient ion diffusivity. To make Mn₃O₄ more applicable, we developed a convenient one-pot synthesis route to fabricate porous hierarchical spherical Mn_3O_4 with in situ coated conductive carbon (C- Mn_3O_4). The C- Mn_3O_4 shows a large capacity and good cycling stability. When assembled into anodes, this material delivered a capacity of 703 mA h g⁻¹ in a 1000 mA g⁻¹ cycling test after 700 cycles with only a 3% capacity decay. Meanwhile, the system provided superior rate performance with capacities of 860, 823, 760, 674, and 570 mA h g⁻¹ at 100, 200, 500, 1000, and 2000 mA g⁻¹, respectively. On the basis of our systematic investigations, we attribute this high electrochemical performance to the carbon reinforced porous hierarchical sphere structure.



KEYWORDS: Mn₃O₄, anode, hierarchical, carbon coating, Li-ion battery

1. INTRODUCTION

Lithium-ion battery (LIB) technology is no doubt an important foundation of our modern lifestyle, considering its dominant place in power storage and supply in the evergrowing-and-upgrading portable electronics and electric vehicle (EV) markets. 1-3 Hence, there is a constant demand for high capacity LIBs. The Ni rich $\text{LiNi}_x\text{Co}_y\text{Mn}_{(1-x-y)}\text{O}_2$ (0 < x, y < 1) LIB cathode materials deliver a capacity of approximately 200 mA h g-1, which is dozens higher than that of the conventional LiCoO₂ material, 4-6 making it the most powerful cathode material reported to date; however, there are safety matters associated with this cathode that have yet to be addressed. Conversely, choices of anode materials are much less restricted.^{7,8} For example, a successful replacement of commercial graphite anodes (372 mA h g⁻¹) with transition metal oxides (TMOs, over 800 mA h g⁻¹) could lead to capacity improvements that would be in the hundreds.

TMOs were first proposed as anodes for LIBs by Poizot et al. in the year 2000. The high capacity of TMOs is considered the benefit of their conversion mechanism, which delivers much higher capacities than those of most intercalation/extrusion cathode materials. Among reported TMOs, Mn₃O₄ is favored by researchers for its low cost, environmental friendliness, and low working potential. ^{1,11-13} The typical voltage window of Mn₃O₄ versus Li/Li⁺ is 0.5-2.0 V. Its theoretical capacity reaches 936 mA h g⁻¹. However, its inferior electroconductibility (10⁻⁷-10⁻⁸ S cm⁻¹) causes poor rate performance. Furthermore, the volume change of Mn₃O₄

during lithiation and delithiation leads to rapid capacity fading. These issues make pure Mn₃O₄ unpractical for scale deployment for LIBs. 9,14,1

Two main strategies are considered effective to address these issues. First, to allow electrons to pass more quickly among Mn₃O₄ particles, different carbon materials or other conductive materials have been combined with Mn₃O₄ to form hybrid structures. 16,17 On the other hand, certain nanostructure designs can promote structural stability against volume change and provide fast ion diffusion pathways. ^{10,18} For example, Yao et al. developed a Mn₃O₄ and conductive carbon matrix. their work, both the capacity and cycle stability of Mn₃O₄ were improved. The reversible specific capacity reached 879 mA h g-1 at a 100 mA g-1 current rate. Mani and teammates prolonged the cycle life of Mn₃O₄ to 9000 cycles with a 20% capacity drop by anchoring Mn₃O₄ particles to graphene.²⁰ Sun and co-workers fabricated self-supported Mn₃O₄@C nanotube arrays via a template-assisted route. Their product showed a splendid rate capability (i.e., reversible capacities of 420, 400, and 375 mA h g⁻¹ at 400/1586, 500/1982, and 600/2380 mA cm⁻²/mA g⁻¹, respectively).²¹ Peng and colleagues succeeded in combining Mn₃O₄ nanoparticles with graphitic nanotubes to reach a discharge capacity over 1000 mA h g-1.22 However, challenges still remain in the preparation of high

Received: May 4, 2019 Accepted: June 24, 2019 Published: June 24, 2019



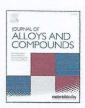
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Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom



Effect of metal ion concentration in precursor solution on structure and electrochemical performance of LiNi_{0.6}Co_{0.2}Mn_{0.2}O₂



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ARTICLE INFO

Article history:
Received 25 June 2018
Received in revised form
13 November 2018
Accepted 15 November 2018
Available online 16 November 2018

Keywords: LiNi_{0.6}Mn_{0.2}Co_{0.2}O₂ Carbonate co-precipitation Metal ion concentration Lithium-ion battery

ABSTRACT

LiNi $_{0.6}$ Mn $_{0.2}$ Co $_{0.2}$ O $_{0.2}$ (622NCM) cathode materials are synthesized by a carbonate co-precipitation process. We decomposed Co and Mn carbonates to form porous CoMnO $_{3}$ precursors, which were impregnated with Ni and Li salts to form final materials. To provide better hosts for the Ni and Li salts, we design the co-precipitation solutions with different total metal ion concentrations to achieve precursors with stable structures and appropriate porosity. We obtained 622NCM with hierarchical and porous properties, which shortened the lithium ion transmission distance and improved the electrolyte infiltration rate and rate capability. Furthermore, our 622NCM cathode materials showed no loss of thermodynamic or electrochemical stability during cycling. The most suitable concentration was determined to be 0.03 mol L⁻¹ and the as-prepared 622NCM (0.03–622NCM) cathode material exhibited good cycle reversibility, a high capacity, and good rate capability, delivering a discharge capacity of 152 mA h g⁻¹ at 0.2 C. A capacity of more than 100 mA h g⁻¹ was achieved at a high rate of 5 C. This study provides guidance for preparing nickel-rich ternary cathode materials for high-performance lithium-ion batteries.

1. Introduction

Lithium-ion batteries (LIBs) are considered to be promising power sources for electric vehicles. However, the performance of commercial LIBs cannot meet growing demand for even higher energies and power densities [1]. Cathode materials have lower energy densities than anode materials, which is a main factor limiting further improvements of LIB performance [2,3]. Thus, further improvements to the energy density of cathode materials are needed. In recent years, the $\text{LiNi}_x\text{Co}_y\text{Mn}_{1-x-y}\text{O}_2$ (NCM) ternary material family has been identified as a promising cathode material candidate for LIBs [4,5] owing to its prominent advantages, such as a high energy density, long cycle lifetime, good security and environment friendliness [6,7]. Among the NCM family, Ni-rich (x > 0.5) materials have a high capacity and low cost, and these features have attracted attention for applications as cathode materials [8-10]. The high discharge capacity of Ni-rich materials is attributed to their relatively high nickel content, which can be explained by the

LiNi_{0.6}Co_{0.2}Mn_{0.2}O₂(622NCM) is a representative nickel-rich ternary cathode material for Li-ion batteries, which has drawn interest since it was first prepared by Cao et al. [15]. 622NCM has a similar crystal structure to those of traditional ternary cathode materials and established hydroxide co-precipitation synthesis processes can be applied to 622NCM. However, the synthesis of stoichiometric NCM materials is regarded as a great challenge owing to the precision required over the experimental conditions, including the solution pH and the need for a protective atmosphere. Recently, Zheng et al. reported a simple carbonate co-precipitation method to prepare 622NCM [16], with more robust reaction conditions, owing to the greater stability of manganese carbonate than that of manganese hydroxide [17]. Moreover, porous structures can be easily formed during calcination of the carbonate precursor. Such structural features benefit lithium ion transport in electrochemical reactions. However, in the carbonate co-precipitation method, nickel salts do not exist in the precursor but are added with lithium during a following impregnation step because of the

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role of redox couples in the NCM materials: The Ni^{2+}/Ni^{4+} redox couple contributes most of the capacity and the Co^{3+}/Co^{4+} redox couple participates in electrochemical reactions and slightly increases the conductivity. The Mn^{4+} ions are electrochemically inactive and stabilize the crystal structure [11–14].

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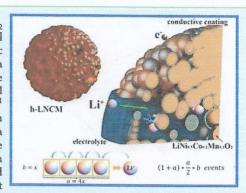
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Compound-Hierarchical-Sphere LiNi_{0.5}Co_{0.2}Mn_{0.3}O₂: Synthesis, Structure, and Electrochemical Characterization

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[†]Beijing Key Laboratory of Environmental Science and Engineering, Beijing Institute of Technology, Beijing 100081, China [‡]Collaborative Innovation Center of Electric Vehicles in Beijing, Beijing 100081, China

ABSTRACT: Compound-hierarchical-sphere-structured LiNi_{0.5}Co_{0.2}Mn_{0.3}O₂ was synthesized to improve the electrochemical performance of this material in lithium-ion battery cathodes. The product was found to have a large specific surface area, good electron and ion conductivities, a stable interface, and a robust nano/microhierarchical structure, all of which improved the rate capability, capacity, and cycling stability of this material. When this material was cycled between 3.0 and 4.3 V, a high discharge capacity of 180.8 mA h g⁻¹ was obtained at 0.2C with 94.0% capacity retention after 100 cycles. In addition, a superior discharge capacity of 148.9 mA h g⁻¹ was observed at a high current density of 1600 mA g⁻¹. This compound-hierarchical-sphere LiNi_{0.5}Co_{0.2}Mn_{0.3}O₂ is readily prepared using our ternary coprecipitation method. We also propose an effector unit theory to explain the enhanced cycling stability of this substance and believe that the present results will assist in the design of cathode materials for lithium-ion batteries.



KEYWORDS: compound-hierarchical-sphere, lithium-ion battery, cathode, LiNi_{0.5}Co_{0.2}Mn_{0.3}O₂, effector unit

1. INTRODUCTION

Clean, high-performance energy storage will play a key role in mitigating environmental concerns associated with present-day energy structures. 1-4 Notably, lithium-ion batteries (LIBs) have already made it possible to reduce road CO2 emissions by replacing combustion engines with battery-powered electric motors. In fact, many modern electronic devices also depend heavily on progress in LIBs and other high-end energy-storage technologies.

Further improvements in LIB technology will require the exploration of high-performance cathode materials. 5-9 In recent years, the LiNi_xCo_yMn_{1-x-y}O₂ family, characterized by high discharge capacities, moderate voltage platforms, low cost, and low environmental toxicity, has garnered interest in research and applied studies. 10-14 As with other members of this family, LiNi_{0.5}Co_{0.2}Mn_{0.3}O₂ adopts a rhombohedral α-NaFeO₂ structure $(R\overline{3}m)$ with alternating $[MO_2]^-$ (M = Ni,Co, and Mn) and Li⁺ layers. Ni²⁺/Ni⁴⁺ and Co³⁺/Co⁴⁺ redox reactions contribute to Li+ transfer in this material, and so the discharge capacity of $\text{LiNi}_{0.5}\text{Co}_{0.2}\text{Mn}_{0.3}\text{O}_2$ is relatively high considering its increased proportion of Ni+. In addition, the Mn4+ proportion in this compound tends to stabilize the crystal structure while reducing cation dissolution and John-Teller distortion. 15,16

Since the discovery of LiNi_{0.5}Co_{0.2}Mn_{0.3}O₂, chemists and material scientists have attempted to optimize its electrochemical performance both by altering the synthesis process to achieve the appropriate crystalline properties and by combining LiNi_{0.5}Co_{0.2}Mn_{0.3}O₂ with various cations or compounds. ^{13,17–22} In our opinion, the best strategy is to In our opinion, the best strategy is to

produce fine crystals in conjunction with nano/microstructural designs that promote electrochemical conversion, such as increased cation ordering and appropriately enlarged surface

In the present work, we developed a facile ternary coprecipitation method for the synthesis of compoundhierarchical-sphere LiNi_{0.5}Co_{0.2}Mn_{0.3}O₂ (h-LNCM). In this process, spherical microparticles are initially prepared via a coprecipitation process. These particles are porous and have a nano/microhierarchical structure as a result of a contraction effect during the oxidative decomposition of the coprecipitate precursors. These structural features are expected to provide a large specific surface area, short lithium-diffusion pathways, fast lithium-transfer channels, and good mechanical strength. In addition, a conductive compound layer is added to provide a stable interface and rapid electron transfer to the particle centers. This elaborate design is illustrated in Figure 1. An effector unit theory was also developed during this study to provide a better understanding of the design of electrode material structures for use in LIBs.

2. MATERIALS AND METHODS

2.1. Materials' Preparation. NiCl₂·6H₂O (99.9%), CoCl₂·6H₂O (99.9%), MnCl₂·4H₂O (99.9%), and NaHCO₃ (99.9%) were purchased from Macklin, Inc. (Shanghai, China), and LiNO3 (99.98%) was purchased from Aladdin (Shanghai, China).

Received: June 15, 2018 Accepted: August 28, 2018 Published: August 28, 2018



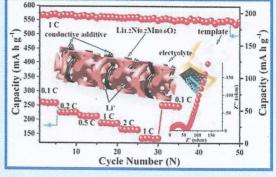
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3D Reticular Li_{1.2}Ni_{0.2}Mn_{0.6}O₂ Cathode Material for Lithium-Ion **Batteries**

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ABSTRACT: In this study, a hard-templating route was developed to synthesize a 3D reticular Li_{1,2}Ni_{0,2}Mn_{0,6}O₂ cathode material using ordered mesoporous silica as the hard template. The synthesized 3D reticular Li_{1.2}Ni_{0.2}Mn_{0.6}O₂ microparticles consisted of two interlaced 3D nanonetworks and a mesopore channel system. When used as the cathode material in a lithium-ion battery, the as-synthesized 3D reticular Li_{1,2}Ni_{0,2}Mn_{0,6}O₂ exhibited remarkably enhanced electrochemical performance, namely, superior rate capability and better cycling stability than those of its bulk counterpart. Specifically, a high discharge capacity of 195.6 mA h g⁻¹ at 1 C with 95.6% capacity retention after 50 cycles was achieved with the 3D reticular $\rm Li_{12}Ni_{02}Mn_{0.6}O_2$. A high discharge capacity of 135.7 mA h g⁻¹ even at a high current of 1000 mA g⁻¹ was also obtained. This excellent electrochemical performance of the 3D reticular Li_{1.2}Ni_{0.2}Mn_{0.6}O₂ is



attributed to its designed structure, which provided nanoscale lithium pathways, large specific surface area, good thermal and mechanical stability, and easy access to the material center.

KEYWORDS: 3D reticular, Li_{1.2}Ni_{0.2}Mn_{0.6}O₂, cathode, lithium-ion battery, hierarchy

1. INTRODUCTION

Cathode material is one of the major components of modern lithium-ion batteries and directly influences the performance and price of the final battery pack. The high cost of current state-of-the-art commercialized lithium-ion batteries is still the main barrier to the storage of energy on the electrical grid to improve grid efficiency and stability while enabling integration of intermittent renewable energy technologies (such as wind and solar) into the baseload supply. Furthermore, user experiences of electric vehicles and portable electronics are rarely free from battery life anxiety.

Recently, the lithium-rich cathode material Li_{1.2}Ni_{0.2}Mn_{0.6}O₂ has attracted much attention for its high specific capacity (ca. 250 mA h g⁻¹), a promising factor for long battery life. $^{2-13}$ It is believed that this layered oxide belongs to the xLi₂MnO₃·(1x)LiMO₂ (M = Mn, Co, or Ni) family, which is composed of two layered α-NaFeO₂-type structures.¹² One of the most interesting features of these lithium-rich materials is that, except for the active LiMO2 (M = Mn, Co, or Ni) phase, they can be charged to a high potential (>4.5 V vs Li), resulting in the electrochemical activation of the Li₂MnO₃ component and a high capacity. Notably, Li_{1.2}Ni_{0.2}Mn_{0.6}O₂ is also relatively cheap and has a low environmental toxicity owing to the absence of cobalt. Along with its intrinsic high capacity, however, Li_{1,2}Ni_{0,2}Mn_{0,6}O₂ suffers from poor rate capacity and modest cycle stability, mainly because of structural rearrangement at its surface during cycling. 12 Specifically, lithium extraction of lithium-rich materials is accompanied by the oxidation of the O_{2n} band (Li₂O) during the initial charge, followed by the irreversible migration of a significant amount of transition-metal ions into the lithium sites.

Many strategies, such as element doping and surface coating, have been adopted to improve the electrochemical performance of lithium-rich layered oxides.^{3,4,14,15} These methods can mitigate the poor electrochemical performance fairly. The recently reported gradient surface Na+ doping method showed an insight by enhancing the kinetics. 16 Meanwhile, reducing the particle size to nanoscale levels has also been proven to be an effective way to improve the rate capability of layered oxides. 13,17,18 Use of nanosized cathode materials shortens lithium-ion diffusion pathways and increases the size of the active surfaces, mainly the (010) lattice planes in the case of α -NaFeO₂ structure layered oxides such as Li_{1,2}Ni_{0,2}Mn_{0,6}O₂; therefore, electrode kinetic issues can be circumvented by switching to nanomaterials. This is an approach with high potential: scientists have been successfully controlling and expanding the properties of materials through nanotechnology in many fields for decades. 19-24 Research also indicates that nanomaterials are usually thermodynamically unstable and tend to agglomerate, which increases the resistance of the final

Received: October 18, 2016 Accepted: December 27, 2016 Published: December 27, 2016

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2018华夏幸福北京马拉松 CFLD BEIJING MARATHON















成绩证书 **CERTIFICATE**

2018.09.16

王乐才 姓名:

WANG LECAL NAME

参赛号码: B3550

BIB No.

性别: 男

GENDER Male

枪声成绩: 3:29:25 **GUN TIME**

净计时成绩:

3:26:37

NET TIME

平均配速: 04:54min/km

AVERAGE PACE

平均时速: 12.25km/h

AVERAGE SPEED

05km 00:25:38/00:25:38

10km 00:49:35/00:23:57 01:12:29/00:22:54 15km

01:35:58/00:23:29 20km

02:00:35/00:24:37 25km

30km 02:24:57/00:24:22

35km 02:50:37/00:25:40

03:16:17/00:25:40 40km

半程净时: 1:40:57 半程枪时: 1:43:45

HALF NET TIME HALF GUN TIME

性别排名 GENDER PLACE

枪声成绩: 3224 23313 人中 名/ **GUN TIME**

净计时成绩: 3042 23313 人中 **NET TIME**

年龄段(18~34岁): 名/ 7423 人中(净计时排名) 938 PLACE BY AGE GROUP

全部选手排名 OVERALL PLACE:

枪声成绩: 3358 28227 人中 **GUN TIME**

净计时成绩: NET TIME

3161 名/ 28227 人中

