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Title(English)	Study on Novel Preparation Process of Olivine Cathode Materials with High Working Potentials and Their Cathode Performance for Lithium Batteries
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論文要約

DISSERTATION OUTLINE

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要約

Thesis Outline

Title:

Study on Novel Preparation Process of Olivine Cathode materials with High Working Potentials and Their Cathode Performance for Lithium Batteries

Outline:

LiCoPO₄ and LiNiPO₄ are expected to be the next generation cathode materials for lithium ion batteries, mainly because of their thermally stable structures, high working potentials and high theoretical capacity (167 mAh g⁻¹). Nevertheless, poor electronic and ionic conductivities of the LiCoPO₄ and LiNiPO₄ cathode materials hinder their applications. To overcome those problems, the LiCoPO₄ and LiNiPO₄ cathode materials were prepared by aerosol and powder technologies in this study. The objective of this study was to improve the electronic and ionic conductivities of LiCoPO₄ and LiNiPO₄ and their electrochemical performance. Furthermore, the electrochemical activity of Ni²⁺/Ni³⁺ redox couple for LiNiPO₄ was also investigated.

Chapter 1 Introduction

The background of energy crisis and the importance of development of renewable energy systems were demonstrated. The rechargeable batteries were introduced as energy storage systems, especially the lithium ion batteries. The cathode materials for lithium ion batteries, particularly LiCoPO₄ and LiNiPO₄, were laid emphasis on, since they could exhibit very high energy densities. The most significant problems of the LiCoPO₄ and LiNiPO₄ cathode materials were revealed. Furthermore, the examples of the recent publications on the LiCoPO₄ and LiNiPO₄ cathode materials were introduced, and then the challenges of the reported synthesis approaches were pointed out in this chapter. Finally, the objective of this study was presented.

Chapter 2 Synthesis and characterization of LiCoPO₄/C composites using various carbon sources

LiCoPO₄/C composites were prepared by a combination of spray pyrolysis and wet ball milling followed by further heat treatment. The effect of various synthesis parameters, such as annealing temperature, rotation speed, content of carbon and various carbon sources on the physical and electrochemical properties of LiCoPO₄/C composites were studied. The effect of crystallinity, carbon content and impurity phase Co₂P on the electrochemical properties of LiCoPO₄/C composites was confirmed. Using polyvinyl pyrrolidone (PVP) as a carbon source, LiCoPO₄/C composites showed a first discharge capacity of 97 mAh g⁻¹ at 0.1C rate.

Chapter 3 Synthesis of spherical nanostructured LiCoPO₄ particles by citric acid assisted spray pyrolysis and their cell performance for lithium batteries

Spherical nanostructured LiCoPO₄ particles (SN-LiCoPO₄) were synthesized by citric acid assisted spray pyrolysis in air atmosphere. The effect of the spray pyrolysis temperature and addition of the citric acid additive in the precursor solution on the physical and electrochemical properties of the LiCoPO₄ samples were investigated. With addition of citric acid in the precursor solution, primary and secondary particle sizes of synthesized SN-LiCoPO₄ were reduced and the specific surface area of the SN-LiCoPO₄ increased significantly. The SN-LiCoPO₄ electrode showed a high discharge capacity of 135 mAh g⁻¹ at 0.05C in a voltage range of 2.0-5.1 V, which reaches to 81% of the theoretical capacity (167 mAh g⁻¹).

Chapter 4 Synthesis and characterization of LiNiPO₄/C composite cathode materials for lithium batteries

LiNiPO₄/C composites by a combination of spray pyrolysis and wet ball milling followed by further heat treatment. The effect of spray pyrolysis temperature, annealing temperature and rotation speed on the physical and electrochemical properties of synthesized LiNiPO₄/C composites were examined. LiNiPO₄/C composites showed a charge capacity of 73 mAh g⁻¹ with a voltage plateau at 5.1 V and an extremely low discharge capacity of 8 mAh g⁻¹ at 0.1C rate.

Chapter 5 Synthesis and characterization of LiNi_{1-x}Co_xPO₄/C (x=0, 0.2, 0.5, 0.8, 1) nanocomposite cathode materials for lithium batteries

LiNi_{1-x}Co_xPO₄/C (x=0, 0.2, 0.5, 0.8, 1) nanocomposites were prepared by a combination of spray pyrolysis and wet ball milling followed by further heat treatment. The effect of cobalt doping content, wet ball milling and carbon coating on the physical and electrochemical properties of synthesized LiNi_{1-x}Co_xPO₄/C nanocomposites were studied. The formation of LiNi_{1-x}Co_xPO₄ solid solutions was confirmed from the linearly change of lattice parameter (*a*, *b*, *c*) with content of Co (*x*). The first discharge capacities of LiNi_{1-x}Co_xPO₄/C nanocomposites were superior to those of bare LiNi_{1-x}Co_xPO₄ samples, due to the reduced particle sizes and introduced conductive carbon. In addition, the electrochemical activity of Ni²⁺/Ni³⁺ redox couple in the LiNi_{0.5}Co_{0.5}PO₄/C nanocomposite electrode was verified from the reduction peak of the cyclic voltammetry curve at 4.7 V.

Chapter 6 Summary

Conclusions based on the above results were summarized.