



Regular Article

Zinc naphthalenedicarboxylate coordination complex: A promising anode material for lithium and sodium-ion batteries with good cycling stability

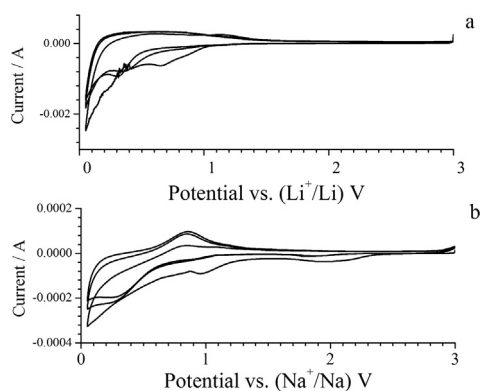


Hailong Fei ^{a,b,*}, Wenjing Feng ^a, Tan Xu ^a

^a College of Chemistry, Fuzhou University, Fuzhou, Fujian 350002, China

^b Key Laboratory of Advanced Energy Materials Chemistry (Ministry of Education), Nankai University, Tianjin 300071, China

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 27 September 2016

Accepted 2 November 2016

Available online 3 November 2016

Keywords:

Dicarboxylate

Anode

Lithium-ion battery

Sodium-ion battery

Dicarboxylic acid

ABSTRACT

It is important to discover new, cheap and environmental friendly electrode materials with high capacity and good cycling stability for lithium and sodium-ion batteries. Zinc 1,4-naphthalenedicarboxylate was firstly found to be stable anode materials for lithium and sodium-ion batteries. The discharge capacity can be up to 468.9 mAh g⁻¹ after 100 cycles at a current density of 100 mA g⁻¹ for lithium-ion batteries, while the second discharge capacity of 320.7 mAh g⁻¹ was achieved as anode materials for sodium-ion batteries. A possible electrochemical reaction mechanism was discussed.

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1. Introduction

With fossil fuels depletion, global warming and environmental pollution, increasing attention has been paid to develop and use

renewable clean energy to lessen greenhouse gas emissions. Lithium-ion batteries with high energy density, low cost and long cycle life are the ideal power source for electric vehicles, which has great significance for lessening greenhouse gas emissions and alleviating the energy crisis [1]. In addition, sodium-ion battery has been brought into focus in large-scale electric energy storage applications for renewable energy and smart grid because of its huge abundant sodium resources and lost cost [2]. Nowadays, great

* Corresponding author at: College of Chemistry, Fuzhou University, Fuzhou, Fujian 350002, China.

E-mail address: hailongfei@fzu.edu.cn (H. Fei).

attentions have been drawn to develop electrode materials for lithium and sodium-ion batteries with high capacity and good cycling stability to meet the need of vehicle power supply in the future.

Conjugated dicarboxylates are promising electrode materials for lithium-ion batteries [3,4]. Recently, metal dicarboxylate coordination polymers has attracted increasing interests in lithium-ion batteries for high capacity and good cycling stability, such as Cu-dicarboxylate-based metal-organic framework $[\text{Cu}_2(\text{C}_8\text{H}_4\text{O}_4)_4]_n$ [5], $\text{CaC}_8\text{H}_4\text{O}_4$ dicarboxylate [6], manganese-based layered coordination polymer $[\text{Mn}-(\text{tfbc})(4,4'\text{-bpy})(\text{H}_2\text{O})_2]$ [7], $\text{Co}_2(\text{OH})_2\text{BDC}$ (BDC $_{1/4}$,1,4-benzenedicarboxylate) [8], a family of transition metal-organic coordination complexes with terephthalate ligands $\text{NiC}_8\text{H}_4\text{O}_4 \cdot 4\text{H}_2\text{O}$ [9] and manganese or cobalt 1,3,5-benzenetricarboxylate [10,11]. Though the element Zinc based ZnO [12], Zn-Sb intermetallic nanowires [13] and zinc stannate [14] have been widely investigated as electrode materials for lithium and sodium-ion batteries. Little attention was paid to test Zn based coordination complex as anode materials for lithium and sodium-ion batteries. $\text{Zn}_4\text{O}(1,3,5\text{-benzenetribenzoate})_2$ [15] exhibited low discharge capacity and poor cycling stability. Metal formate $\text{Zn}_3(\text{HCOO})_6$ with diamondoid structure was obtained with a discharge capacity up to 560 mAh g^{-1} (9.6 mol of Li) as with 60 cycles at a current density of 60 mA g^{-1} , which is attributed to the reversible formation or regeneration of FOR1 MOF during the cycling and electrochemical alloying-dealloying process of Zn [16]. Though transition metal polypyrrole, polythiophene and pyridinedicarboxylate coordination complex shows a high discharge capacity and good cycling stability [17–21]. Few attentions were drawn to transition metal dicarboxylate coordination polymer with heteroatom organic ring [22]. As far as we know, few metal dicarboxylates were investigated as electrode materials for sodium-ion batteries. Herein, we prepared a new zinc 1,4-naphthalenedicarboxylate coordination polymer. It is firstly found to be a kind of high-capacity anode material for lithium and sodium-ion batteries with good cycling performance.

2. Material and methods

All chemicals are commercially available and used as received. In a typical procedure, equal mmol 1,4-naphthalenedicarboxylic acid and zinc nitrate was added to the mixed solution of water and ethanol and stirred at room temperature for 2 h. After that, the mixture was transferred to a 50-ml Teflon-lined stainless autoclave, sealed, kept at 200°C for 24 h, cooled to room temperature, washed with absolute alcohol and dried at 70°C for 12 h.

The morphological characteristics of the as-synthesized materials were observed with a Hitachi S-4800 field emission scanning electron microscope (SEM). X-ray diffraction (XRD) patterns were recorded on a diffractometer (CoK α , PANalytical, and X'Pert). The FT-IR spectra were recorded on an America Thermo Fisher Scientific Nicolet 6700 spectrometer. A Land CT2001A battery tester was used to measure the electrode activities at room temperature. A Varo EL elemental analyzer from German Elementary Analyses System GmbH and Optima 8000 ICP-OES from American Perkin-Elmer were used for elemental analysis of H, C, N and Zn. Thermal analysis measurements were performed using a German Netzsch DIL402C Analyzer. Cyclic voltammetry (CV) experiments were performed with a Chi660c electrochemical workstation at a scan rate of 1 mV s^{-1} .

The coordination polymer was tested as anode materials for lithium and sodium-ion batteries. The composite of negative electrode material was consisted of the active material, a conductive material (super-pure carbon) and binder polyvinylidene difluoride in a weight ratio of 6/3/1. The electrolyte was 1 M LiPF_6 in a 1/1/1 (volume ratio) mixture of ethylene carbonate (EC), propylene

carbonate (PC) and dimethyl carbonate (DMC). The Li metal was used as the counter electrode. For sodium-ion batteries, the Na metal was used as the counter electrode. The electrolyte was 1 M NaClO_4 in a 1/1 (volume ratio) mixture of ethylene carbonate (EC) and propylene carbonate (PC) with the addition of 5% fluoroethylene carbonate (FEC). The cells were charged and discharged between 0.05 and 3.0 V limit.

3. Results and discussion

X-ray diffraction was performed to identify the crystalline structure, as shown in Fig. 1a. The X-ray diffraction patterns show that zinc 1,4-naphthalenedicarboxylate is crystalline (Fig. 1a). The strong peak at 6.99° implied that is phase-pure. However, it cannot be ascribed to the known materials in the XRD database. It is not single crystal, thus it is difficult to manifest the main diffraction peaks and structure.

The structure information of the as-synthesized coordination polymer was provided by FT-IR spectrum (Fig. 2a). The IR peaks

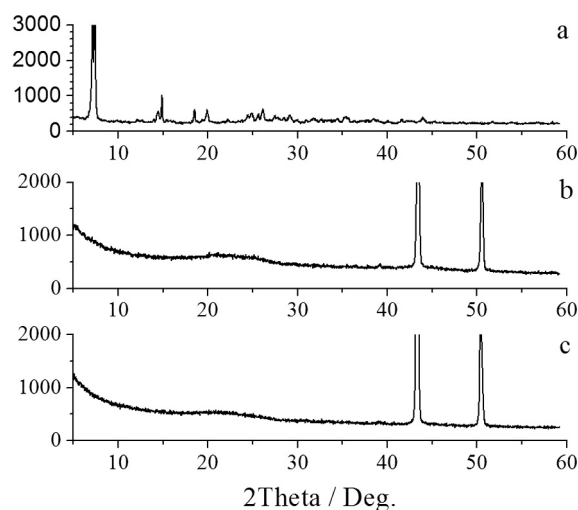


Fig. 1. Wide angle XRD patterns of (a) Zn 1,4-naphthalenedicarboxylate and its first discharge and charge products (b and c).

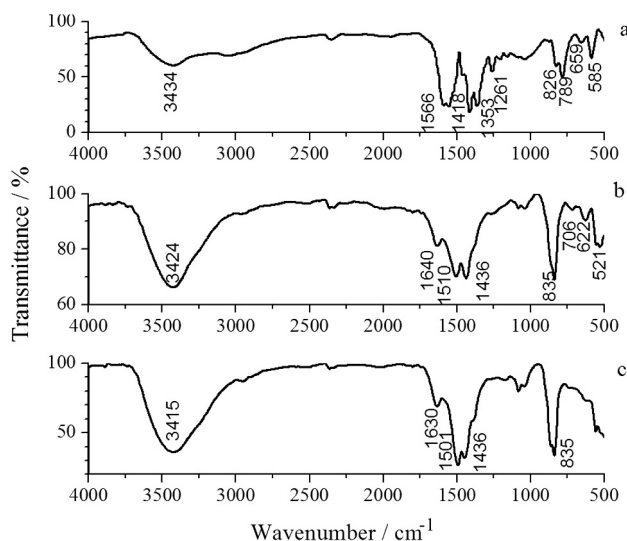


Fig. 2. FT-IR spectra of (a) Zn 1,4-naphthalenedicarboxylate and its first discharge and charge products (b and c).

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