

β -Bi₂O₃: An underlying negative electrode material obeyed electrode potential over electrochemical energy storage device



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ABSTRACT

An underlying negative electrode material of β -Bi₂O₃ has been designed according to the electrode potential, and synthesized via the combination of a similar hydrothermal method and subsequent annealing treatment. The electrochemical evaluation shows that the metastable phase of β -Bi₂O₃ electrode possessing a wide potential window between -1.5 V to 1.5 V in neutral electrolyte manifests satisfying capacity of 871.2 C g⁻¹, superior specific energy of 266 Wh kg⁻¹, outstanding rate capability and excellent cycling stability, which confirms this design and provides a feasible method for later design of negative electrode materials. Based on the morphological characteristics and electrochemical measurements, a possible mechanism is proposed that it'll more conducive to electrochemical behavior for particles owning smooth surface. Also, as an electrochemical application, an electrochemical energy storage device has been assembled, where MnO₂ and the β -Bi₂O₃ acted as the positive and the negative electrodes, respectively, and the specific energy of 32.4 Wh kg⁻¹ is demonstrated at a cell voltage between 0 V to 1.8 V, exhibiting a high energy density and stable power characteristic, which revealed that such a material is promising in electrochemical energy storage applications.

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1. Introduction

Most commercial electrochemical energy storage devices are using C-based materials as electrodes, store energy via a charge separation at the electrode/electrolyte interface, and this class of devices is named electrochemical double layer capacitors [1]. The capacity of such electrochemical capacitors is limited to about 250 F g⁻¹ in aqueous electrolytes for high surface area C-based materials [2].

To overcome this limitation, new breeds of electrochemical energy storage devices possessing outstanding electrochemical performance have been developed [3–6]. Main conclusions about the electrode materials, in general, having a smaller potential window than that of C or C-based materials, are the lack of electronic conductivity and rate capability compared to the C-based materials [7–13], which severely limits the energy density of the electrodes.

A tremendous amount of work has been developed on Ni-, Co-, and Mn-based compounds positive electrode materials obeyed their potential windows [14–26], owing to the first-class

electrochemical properties, especially, the rate capability after improved electronic conductivity, which led to almost all of existing asymmetric electrochemical energy storage devices are using the type that these electrode materials mentioned above acted as the positive electrode materials and C or C-based materials were still only used as the negative electrode materials. But the consequence is that dynamics performance between the positive electrode materials and the negative electrode materials is scarcely matching.

Rather than improving the performance of only several kinds of positive electrode materials or developing new kinds of positive electrode materials, it appears to be more significant to design or develop the novel negative electrode materials. Regrettably, there's little researchers focused on developing or designing new negative electrode materials except Fe-based compounds [27–30].

Here begins a simple idea with the electrochemical electrode chemical reaction: $\text{BiO}^+ + 2\text{H}_2\text{O} = \text{Bi}_2\text{O}_4 + 2\text{e}^- + 4\text{H}^+$; V (versus Standard Hydrogen Electrode) = -1.59 V [31]. It should be point out that the electrode potential V in the reaction is -1.59 V, which illuminates that such class of Bi-based compounds stands a chance to become an underlying negative electrode material for electrochemical energy storage devices. Those Bi-based compounds which meet these conditions in the reaction are the compounds possessing an oxidation state of +3 for Bi element. In the

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circumstances, Bi₂O₃ which is low-cost and simple to synthesize is a preferred option.

In fact, there are several reports, such as Zheng's [32], Qu's [33], Nithya's [34], and Senthilkumar's [35] works, about Bi-based compounds acted as the electrode materials but not exactly the negative electrode materials for electrochemical energy storage devices, which shows that our guess, anyhow, is doubtless, and also greatly inspire us to design and develop it.

In this paper, a new breed underlying negative electrode material of Bi₂O₃ is designed according to the electrode potential, developed obeying the electrode chemical reaction, and synthesized via the combination of a similar hydrothermal method and subsequent annealing treatment. The electrochemical evaluation shows that the metastable phase β -Bi₂O₃ electrode possessing a wide potential window between -1.5 V to 1.5 V in neutral electrolyte manifests satisfying capacity of 871.2 C g⁻¹, outstanding rate capability and excellent cycling stability, which confirms our design and provides a feasible method for later design of electrode materials.

Based on the morphological characteristics and electrochemical measurements, a possible mechanism is proposed that it'll more conducive to electrochemical behavior for particles owning smooth surface. Also, an electrochemical energy storage device has been assembled, where MnO₂ and β -Bi₂O₃ acted as positive and negative electrodes, respectively, and the specific energy of 32.4 Wh kg⁻¹ is demonstrated at a cell voltage between 0 V to 1.8 V, exhibiting a high energy density and stable power characteristic, which revealed that such a material is promising in electrochemical energy storage applications.

2. Experimental

2.1. Materials synthesis

Analytical grade Bi(NO₃)₃·5H₂O and N, N-dimethyl formamide (DMF) were purchased from Sinopharm Chemical Reagent Co. Ltd.

Firstly, 2 g of Bi(NO₃)₃·5H₂O were dissolved in 50 mL DMF. Secondly, the obtained solution was transferred into Teflon-lined stainless steel autoclave at 180 °C for 12 h. Thirdly, filtration and vacuum drying at 60 °C for 12 h to obtain the precursor. Fourthly, the precursor was annealed in air 300 °C for 5 h to obtain the slippery β -Bi₂O₃ nanohunks. Commercial Bi powder was annealed in air 300 °C for 5 h to obtain the amorphous β -Bi₂O₃ nanoparticles.

2.2. Structure characterization

Crystallite structure was determined via X-ray diffraction (XRD) using a Rigaku D/MAX2400 diffractometer with the Cu K α radiation (wavelength $\lambda=0.15418$ nm) operating at 40 kV and 60 mA. Microstructures and morphologies were characterized via field emission JEOL JSM-6701F scanning electron microscope (SEM) and JEOL JEM-2010 transmission electron microscope (TEM).

2.3. Electrodes preparation

80 wt.% of the active material was mixed with 7.5 wt.% of acetylene black and 7.5 wt.% of conducting graphite in an agate mortar until a homogeneous black powder was obtained. 5 wt.% of polytetrafluoroethylene was added together with a few drops of ethanol. The resulting paste was pressed at 10 MPa into an open-cell nickel foam, dried at 80 °C for 12 h. Every electrode possesses a geometric surface area of 1 cm².

2.4. Electrochemical evaluation

Electrochemical measurements were carried out using Chenhua CHI660C electrochemical working station in a three-electrode cell at about 20 °C. A platinum electrode and a saturated calomel electrode served as the counter electrode and the reference electrode, respectively. The electrolyte was 1 mol L⁻¹ aqueous Na₂SO₄ solution.

The corresponding capacity was calculated from the following equations:

$$C_m = \frac{I \cdot \Delta t}{m} \quad (1)$$

And, the energy density E was calculated from the following equation:

$$E = C_m \cdot \Delta V \quad (2)$$

Also, the power density P which was determined from the constant current charge/discharge cycles is as follows:

$$P = \frac{I \cdot \Delta V}{m} \quad (3)$$

where C_m (C g⁻¹) is the capacity, I (A) is discharge current, Δt (s) is the discharge time, ΔV (V) represents the average potential, and m (g) is the mass of the materials, E (Wh kg⁻¹) is the energy density, P (W kg⁻¹) is the power density.

2.5. Electrochemical energy storage devices

The electrochemical energy storage devices consisted of two electrically isolated electrodes: MnO₂ electrode as the positive electrode and the β -Bi₂O₃ electrode as the negative electrode joined together by a porous non-woven cloth separator soaked in 1 mol L⁻¹ aqueous Na₂SO₄ solution. Each electrode has a geometric surface area of 1 cm² as well.

For the devices, the charge balance follows the relationship:

$$Q_+ \equiv Q_- \quad (4)$$

where Q_+ and Q_- represent the charge stored in positive electrode and negative electrode, respectively. The Q of each electrode depends on the capacity (C), the potential range (ΔV) of the charge/discharge tests, and the mass of the electrode (m) according to the following equation:

$$Q = C_m \cdot m \quad (5)$$

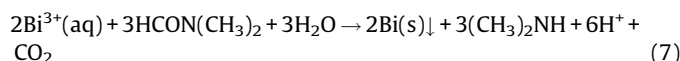
When $Q_+ = Q_-$, the ratio of the masses of the positive electrode (m_+) and negative electrode (m_-) will follow the equation:

$$\frac{m_+}{m_-} = \frac{C_{m-}}{C_{m+}} \quad (6)$$

3. Results and discussion

3.1. Materials structures

Here, the chemical reactions in the process of materials synthesis are as follows:



It should be noted that the DMF acted as an electron-donating group in reaction (8) and provided electron, while the Bi³⁺(aq)

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