



Silver nitrate nanosheet supported on porous carbon three-dimensional substrate as cathode material and its lithium storage mechanism



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ABSTRACT

In this paper, we present the direct application of AgNO_3 as a novel host material for lithium storage. Fabricated as cathode material, commercial AgNO_3 particles show an initial charge capacity at 157.1 mAh g^{-1} , and retain a capacity of only 11.5 mAh g^{-1} after 100 cycles. By preparing AgNO_3 nanosheets supported on conductive carbon three-dimensional substrate, enhanced lithium storage performance can be achieved. As-prepared AgNO_3 nanosheets exhibit the charge capacity at 141.7 mAh g^{-1} in the first cycle, and maintain at 82.0 mAh g^{-1} after 100 cycles. The three-dimensional carbon substrate with considerable strength and tenacity supporting AgNO_3 nanosheets presents fast electron/ion transport, large electroactive surface area, and excellent structural stability. The reaction mechanism of AgNO_3 with Li is also studied by ex situ and in situ techniques during the initial cycle. It can be concluded that lithium storage process of AgNO_3 is associated with a quasi-reversible electrochemical conversion reaction. During the discharge process, the electrochemical reaction of AgNO_3 with Li results in the formation of Ag and LiNO_3 . In the reverse charge process, AgNO_3 can be re-generated by its conversion reaction. Therefore, it is anticipated that AgNO_3 nanosheets can be a probable cathode candidate for lithium-ion batteries.

1. Introduction

As one of the most significant energy storage systems, lithium-ion batteries (LIBs) play an increasing important role in portable electronic devices and electrical vehicles due to their high performance compared with other battery systems [1,2]. In the meantime, the huge demand for electronic devices, electrical vehicles and large energy conversion equipment requires batteries capable of high energy density, low cost and excellent capacity retention [3,4]. To meet the high demand of the above-mentioned batteries, one of the most important challenges is to develop electrode materials (including cathode materials and anode materials) with high capacity as well as better cycling performance [5–7]. Nowadays, the commercialized electrode materials are still limited to a few highly crystalline ones (like LiCoO_2 and graphite) and the traditional anode materials (for instance, graphite, oxides, and sulfides) generally possess higher theoretical capacity as compared to cathode materials (LiCoO_2 and LiFePO_4) [8,9]. Hence, the successful investigation of advanced cathode materials can be a breakthrough in LIBs as they are the primary limiting factor that hinders LIBs from realizing their full potential in terms of energy density [9]. Considering

the modern cathode materials (V_2O_5 , $\text{Li}_3\text{V}_2(\text{PO}_4)_3$ and transition metal fluorides) mostly face the problems of low rechargeable capacity and inferior ion conductivity [10,11]. Therefore, it is of great demand to develop new cathode materials with high capacity and superior cycling performance to satisfy the growing battery markets.

In recent years, silver vanadium oxides ($\text{Ag}_2\text{V}_4\text{O}_{11}$ and AgVO_3) have been attracted great attention among various cathode materials due to their high specific capacity, high energy density and long-term stability [12,13]. However, the reduction of Ag^+ during the charge/discharge process will cause the structure collapse of the silver vanadium oxides, and thus leading to the poor cycling life and rate capability [13–15]. To address above problems, a large number of methods, such as surface modification [16–18] or nano-structure [19–28] have been applied to restricts the particles from collapse and enhance the electronic conductivity for the sake of improving the cycling performance of silver vanadium oxides. Especially for nano-structure, it not only can shorten the diffusion pathway but also can inhibit volume change during the charging/discharging process compared to bulk materials. For instance, G.Y. Huang et al. reported the Co_3O_4 nanocubes exhibited high initial discharge capacities (1298 mAh g^{-1} at 0.1 C and 1041 mAh g^{-1} at 1 C)

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and impressive rate capability, and excellent capacity retention [22]. Y. Chen et al. reported $\text{Li}_3\text{V}_2(\text{PO}_4)_3/\text{C}$ nanosheets exhibited high discharge specific capacity of 130 mAh g^{-1} in the first cycle and the capacity retention is almost 100% after 100 cycles [27]. From the above research papers, it is concluded that nano-structure materials deliver superior electrochemical prosperities compared with their previous bare particles.

In the search of advanced electrode material, we find that bare AgNO_3 has the potential of being a promising cathode material due to its high working potential (3.55 V). No report can be found about the direct application of AgNO_3 as energy storage material for rechargeable batteries. In addition to this, we adopt a simple ultrasonic assisted solution approach to fabricate an electrode material with AgNO_3 nanosheets. Furthermore, porous carbon substrate with three-dimensional network structure is used as current collector to support AgNO_3 nanosheets, thus leading to hybrid structure and considerable strength, which will fasten lithium ion and electron transportation and buffers the volume change during charge/discharge process [29–31]. Hence, AgNO_3 nanosheets can be a potential cathode candidate for LIBs.

2. Experimental

Porous Ni foam was used as the hard template to prepare porous carbon substrate in this work as described by H.X. Ji [32]. It was cut into discs and then coated by carbon via chemical vapor deposition method using acetylene as carbon source. The resulting substrate was cleaned with $\text{Fe}(\text{NO}_3)_3$ and HCl solution in an ultrasound bath for 20 h before being rinsed with absolute ethanol. The cleaning process is aimed to remove the Ni substrate. The detailed preparation processes, as well as the battery-assembling, are schematically shown in Fig. 1. The AgNO_3 nanosheets were synthesized by an ultrasonic assisted solution approach. First, 0.50 g AgNO_3 was dissolved in 50 mL ethanol to

obtain a uniform and transparent solution after constant stirring for 1 h. Then, the transparent solution was kept for magnetic stirring and ultrasonically handled for 8 min every 4 h. After magnetic stirring for 48 h, the solution was transferred into vacuum oven at 50°C for recrystallization. The schematic fabrication process is depicted in Fig. 2. All the chemical reagents used were of analytical reagent grade, purchased from Guanghua Chemical Reagent Shantou Co. Ltd. in China and used without further purification.

To examine the electrochemical properties, active substance, acetylene conductive black, and polyvinylidene fluoride with a ratio of 87:5:8 were mixed to prepare working electrode, which was dried in a vacuum oven at 60°C for 12 h prior to cell assembling. The Li/AgNO_3 cells were assembled in an Ar-filled glove box using lithium metal foil as the counter/reference electrode. Galvanostatic lithium storage capacities were measured by cycling the half cells in the voltage range of 2.0–4.5 V. The battery performances were tested on multi-channel LANHE battery test system at room temperature. Cyclic voltammogram (CV) at a scan rate of 0.1 mV s^{-1} and electrochemical impedance spectroscopy (EIS) within the frequency range from 100 KHz to 0.01 Hz were carried out on a Bio-Logic VSP-300 potentiostat.

The surface morphology and micro-structure of AgNO_3 nanosheets and particles were evaluated by Hitachi S3400 scanning electron microscopy (SEM). X-ray photoelectron spectroscopy (XPS) was carried out using a focused and monochromized Al K α radiation with a Kratos Axis Ultra spectrometer. To investigate the phase structure of the obtained products, X-ray diffraction (XRD) patterns were recorded by a Bruker AXS D8 Focus power X-ray diffractometer with a Cu K α ($\lambda = 0.15406 \text{ nm}$) radiation source. The samples were scanned with a 2θ range from 10 to 60° , a step size of 0.1° and a count time of 1 s. The in situ X-ray electrochemical cells were redesigned from traditional simulated cells and using Be disc as X-ray transmission window, which also has been discussed in our previous papers [33,34]. During the in

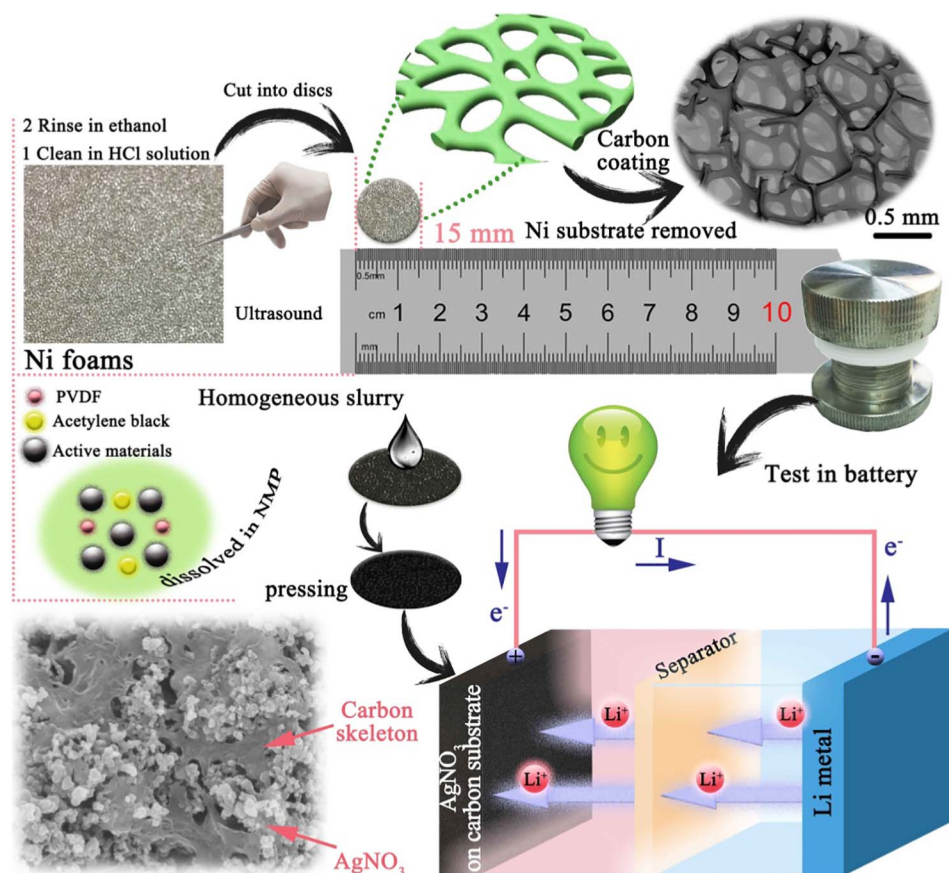


Fig. 1. Schematic fabrication process of AgNO_3 porous carbon substrate and its working model in LIBs.

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