



Chemically modified morphologies of vanadium pentoxide as superior cathode material for lithium ion battery



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ABSTRACT

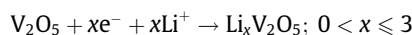
Different morphologies of vanadium pentoxide (V_2O_5) have been synthesized via a facile, eco-friendly, bottom-up and self-assembly approach using hydrothermal process at low temperature for the first time. The as-prepared V_2O_5 plate-like structure shows excellent lithium storage and rate capability compared to tube-like structure and commercial V_2O_5 powder. The specific capacity of V_2O_5 plate can reach 470 mA h g^{-1} at the first cycle with a current density, 17 mA g^{-1} (about 0.05 C) and $110\text{--}120 \text{ mA h g}^{-1}$ at high current density, 1360 mA g^{-1} (about 13 C). The gravimetric energy densities of the plate-like V_2O_5 structure can achieve 1073 W h kg^{-1} and 722 W h kg^{-1} with the current densities of 51 mA g^{-1} and 680 mA g^{-1} , respectively.

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1. Introduction

Emergent need for sustainable energy (creation, alteration, storage and retrieval, etc.) with the cumulative problems of global warming has strengthened the necessity to develop new and more proficient means of acquiring and storing energy. The sustainable energy must be efficiently stored for the future use in devices and vehicles. Li ion battery is one of the most promising candidates for the energy storage systems owing to its outstanding electrochemical performance with high specific capacity and energy density. It is more environmental friendly comparing to the lead acid battery and has higher power density than that of NiH_2 battery used in industry. However, a strong demand in high capacity storage, rate capability and energy density urge the improvement of Li ion battery. Commonly used cathode materials, lithium iron phosphate ($LiFePO_4$), lithium cobalt oxide ($LiCoO_2$), lithium manganese oxide ($LiMn_2O_4$), lithium nickel oxide ($LiNiO_2$), and lithium cobalt manganese nickel oxide (NCMO) possess the specific capacity within a range of $120\text{--}160 \text{ mA h g}^{-1}$. (The specific capacity of some material such as NMC can reach around 200 mA h g^{-1} depending on C-rates) [1–5]. However, these values might not be enough for the future use in the energy storage and vehicles.

Vanadium oxide, especially vanadium pentoxide (V_2O_5) with a layer structure, one of the most potential cathode materials, has been extensively investigated due to its low cost, abundance and its high theoretical capacity that highly depends on its structural phase modifications clearly manifested as plateaus during the charge and discharge cycling. Typically the electrochemical reaction during Li intercalation into V_2O_5 proceeds as:



The α -phase of $Li_xV_2O_5$ exists for $x < 0.01$, ϵ -phase for $0.35 < x < 0.7$, δ -phase for $0.7 < x < 1$ and Υ -phase for $1 < x < 2$. Further insertion of the third lithium into V_2O_5 leads to the irreversible formation of ω -phase ($2 < x < 3$) $Li_xV_2O_5$ with a rock salt type structure. (294 mA h g^{-1} with 2 Li ions intercalation/de-intercalation per unit formula; 440 mA h g^{-1} with 3 Li ions intercalation/de-intercalation per unit formula) [6–11]. Even only one Li ion is intercalated/de-intercalated into V_2O_5 structure, the relatively high capacity can be achieved comparing to the commercialized cathode materials such as $LiCoO_2$ (140 mA h g^{-1}) and $LiMn_2O_4$ (146 mA h g^{-1}) [12–14].

Fabrication of nanostructures in a controlled way with desired composition and geometries inheriting superior or even unusual physico-electro-chemical functionalities is the issue that envisages tremendous efforts. The hierarchical structures or various morphologies of metal oxide would inherit the superior characteristics of the micro/nano building block and obtain additional benefits

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simultaneously from its secondary structure [15–19]. The present study reports a facile, environmental friendly and high yield hydrothermal process to synthesize V_2O_5 with various morphologies, and the schematic morphological change of vanadium pentoxide is shown in Fig. 1. It is revealed that the micro/nanostructures of synthetic V_2O_5 under critical hydrothermal treatment at 180 °C depends strongly on the pH value of the solution, e.g. V_2O_5 tubes and plates can be obtained by adjusting the pH values between 2–3 and 3–4 respectively.

2. Experimental method

2.1. The synthesis of vanadium pentoxide with different morphologies

To fabricate different morphologies of V_2O_5 , ammonium metavanadate (0.5 g) was dissolved in deionized water (50 ml) while stirring at 60 °C to form a clear light yellow solution. Concentrated (60–78%) Nitric acid was added drop-wise to the solution while stirring at the same temperature until the final pH of the solution reaches between 2 and 4. The pH of the system can be easily controlled depending on the desired morphology i.e. 2–3 for tube-like morphology and 3–4 for plate like morphology. As prepared clear orange solution was transferred into a Teflon bottle with a stainless steel shell sealed and maintained at the temperature of 180 °C for 24 h. After the reaction was finished, the reaction mixture was cooled to room temperature. The resulting solid products were washed with distilled water and

ethanol to remove any ions possibly remaining in the final products and then dried at 60 °C in vacuum oven overnight. To get pure V_2O_5 the obtained product were calcinated at 300 °C for 2 h in air.

2.2. Cell preparation and test

For the preparation of the lithium ion battery cathodes, 10 wt% Super P was first mixed with 10 wt% PVDF in N-methyl-2-pyrrolidone (NMP) followed by the addition of 80 wt% of the active material as-synthesized V_2O_5 with various morphologies, and all were mixed with stainless steel balls at 400 rpm for 15 min. The resultant slurry, pasted on an Al foil, was dried at 110 °C for 4 h. The coin cells (2032) then were assembled in an argon-filled glove box using a lithium foil as the anode, Celgard 2325 as the separator, and 1 M $LiPF_6$ dissolved in ethyl methyl carbonate (EMC), dimethyl carbonate (DMC) and vinylene carbonate (VC) with a volume ratio of 1:1:1 as the electrolyte. Cells were tested at ambient environment. The testing voltage in the constant current mode was in a range of 1.75–4 V.

3. Results and discussion

3.1. Scanning and Transmission Electron Microscopy (SEM/TEM)

Fig. 2 displays SEM and TEM images of the as-synthesized products obtained at different pH-values. When the pH of the system is adjusted to 3–4, thick plate structures were obtained as shown in

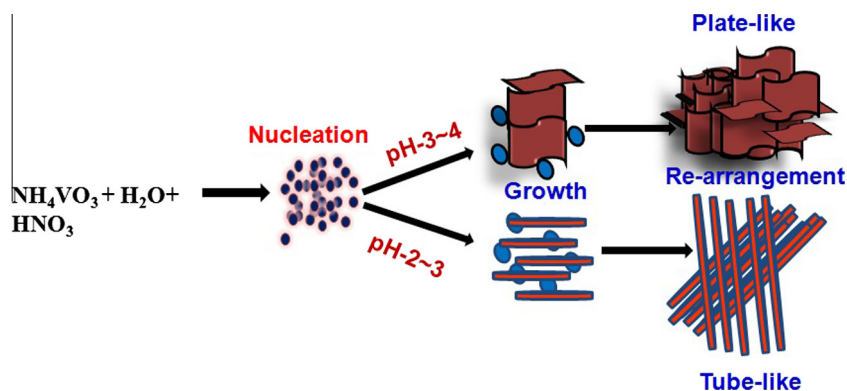


Fig. 1. The schematic morphological change of vanadium pentoxide.

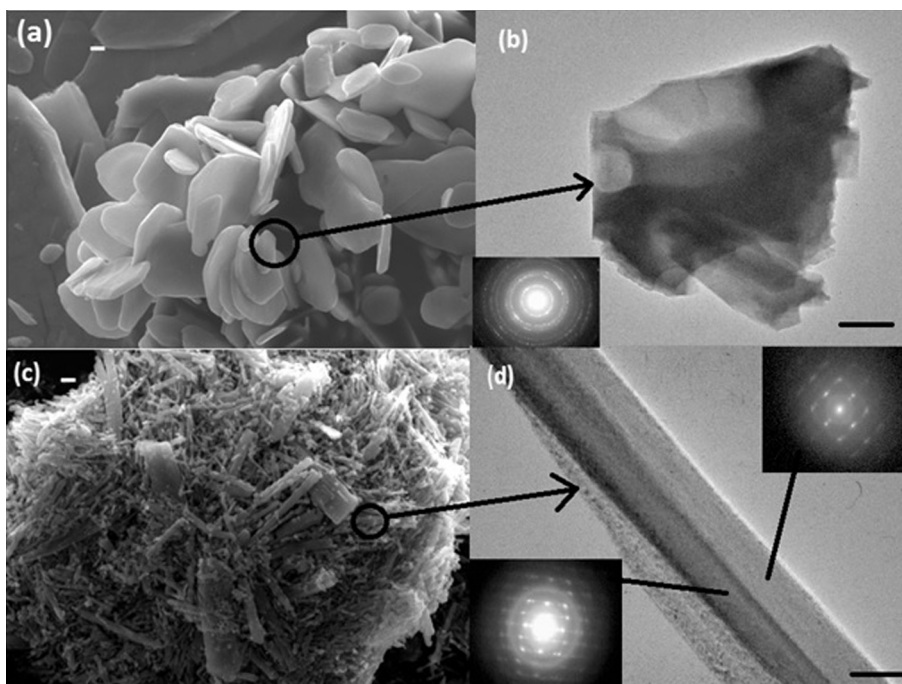


Fig. 2. SEM (left) and TEM (right) images of V_2O_5 plates (a and b at pH 2–3; the scale bar is 1 μ m) and tubes (c and d at pH 3–4; the scale bars are 1 μ m and 50 nm, respectively). The SAED patterns of V_2O_5 plates and tubes structures are shown left-down of (b) and right-up and left-down of (d) respectively.

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