



Adjusting electrode initial potential to obtain high-performance asymmetric supercapacitor based on porous vanadium pentoxide nanotubes and activated carbon nanorods



Zongyuan Lin ^{a, b}, Xingbin Yan ^{b, *}, Junwei Lang ^b, Rutao Wang ^b, Ling-Bin Kong ^{a, *}

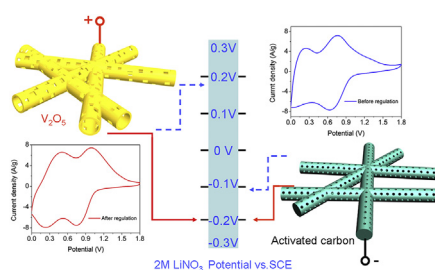
^a State Key Laboratory of Advanced Processing and Recycling of Non-ferrous Metals, School of Materials Science and Engineering, Lanzhou University of Technology, Lanzhou 730050, PR China

^b Laboratory of Clean Energy Chemistry and Materials, State Key Laboratory of Solid Lubrication, Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences, Lanzhou 730000, PR China

HIGHLIGHTS

- Adjusting electrode potential is used to improve the electrochemical property.
- Porous V_2O_5 nanotubes and activated carbon nanorods are used to build asymmetric supercapacitor.
- The $V_2O_5//C$ supercapacitor exhibits excellent cycle stability, and high energy and power densities.

GRAPHICAL ABSTRACT



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ABSTRACT

In this work, a high-performance asymmetric supercapacitor has been developed by using porous vanadium pentoxide (V_2O_5) nanotubes as positive electrode and activated carbon nanorods as negative electrode in an aqueous 2 M $LiNO_3$ electrolyte. To maximize the energy density of the asymmetric supercapacitor, the initial potentials of work electrodes are tuned to different values (0 V, -0.1 V, -0.2 V, and -0.3 V vs. SCE), and the influence of the electrode potential on the electrochemical properties of the obtained asymmetric supercapacitor has been investigated in depth. The results show that -0.2 V is the optimal initial electrode potential. At this initial electrode potential, the built $V_2O_5//C$ asymmetric supercapacitor could be cycled reversibly in the voltage region of 0–1.8 V, and exhibits high energy and power density (46.35 Wh kg^{-1} at 1.8 kW kg^{-1} and 18 kW kg^{-1} at 28.25 Wh kg^{-1}). Furthermore, the supercapacitor shows excellent cycling stability, with an almost 100% specific capacitance retention after 10,000 cycles. The satisfactory results demonstrate that the adjusting of electrode potential is a very effective method to improve the electrochemical performance of asymmetric supercapacitors.

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1. Introduction

Supercapacitors (also called electrochemical capacitors) with

high power density, excellent cycle life and fast charge–discharge ability, have attracted a great deal of attention for the applications in portable electronics and hybrid electric vehicles [1–4]. Depend on the charge storage mechanism supercapacitors can be distinguished as the following types: electrochemical double layer capacitors (EDLCs) and pseudo-capacitors. A typical EDLC is charged

* Corresponding authors.

E-mail addresses: xbyan@licp.cas.cn (X. Yan), longjb@lut.edu (L.-B. Kong).

by the physisorption of electrolyte ions on the electrode materials, but the capacitance occurs in the latter type capacitor is from the underpotential deposition, surface redox reactions and ion intercalation [1,5–7]. Although the specific capacitance of pseudo-capacitors is higher than that of the EDLCs [8], the pseudo-capacitors still suffer from a lower energy density than batteries [9]. To surmount the limit of energy density, various asymmetric supercapacitors are developed by constructing a battery-like Faradic electrode and a capacitive electrode to obtain a higher operating voltage [10–12].

Up to now, considerable research efforts have been devoted to the various asymmetric capacitor systems by assembling different electrode materials such as carbon, transition metal oxides, metal hydroxides, metal nitrides and conductive polymers [3,4,13–18]. Among them, the association of a pseudo-capacitive material and a carbon material both having the high capacitance is the mainstream research direction. For example, a series of asymmetric supercapacitors [19], such as RuO₂//AC (activated carbon) [20], Ni(OH)₂//AC [21], NiO//graphene [22], Co(OH)₂//AC [23], MnO₂//AC [24] have been widely reported, and they all exhibit an increased working potential window and obviously improved energy density compared with aqueous symmetric supercapacitors. However, such a simple constructing method cannot totally fulfill the demand of the energy storage.

In fact, electrode materials with high capacitance and wide operation potential window can not enough ensure the maximized energy density for asymmetric supercapacitors, because a polarization, hydrogen evolution or oxygen evolution occur, when the asymmetric supercapacitor tested at inappropriate electrode initial potential. It is because that electrode materials and the constructing technology are both key factors of building a high-performance asymmetric supercapacitor. Recently, Zakhidiv et al. changed the initial potential of carbon nanotubes (CNTs) electrode to a new electrochemical potential by injecting charge in an aqueous electrolyte, and they found that the new electrochemical potential of CNT electrode could be retained after dipping it back [25]. Also, Cheng et al. assembled an AC symmetric supercapacitor, and the energy density of the symmetric supercapacitor was improved by injecting charge to tune carbon electrode potential [26]. The result proved that the AC symmetric supercapacitor could work in a wide potential region in theory, but the technology of adjusting electrode initial potential makes it come true.

Herein, we construct an asymmetric supercapacitor by using porous V₂O₅ nanotubes as the positive electrode and activated carbon nanorods as the negative electrode in a 2 M LiNO₃ aqueous electrolyte. On this basic, we verify the effectiveness of adjusting the initial potentials of both electrodes to maximize the energy density of the asymmetric supercapacitor. Here, the porous V₂O₅ nanotubes are chosen as the positive electrode material, which is due to that they have a high specific capacitance and a wide working potential region [27–30]. Meanwhile, the activated carbon nanorods have superior electrochemical features in our previous works [31,32]. In this study, when the initial electrode potentials of two electrodes are adjusted to –0.2 V (vs. SCE), the built V₂O₅//C asymmetric supercapacitor has the maximum energy density of 46.35 Wh kg^{–1} at the power density of 1.8 kW kg^{–1}. Moreover, this device shows excellent cycling stability for 10,000 cycles without any decay. To our best knowledge, this is the first time that both Faradic and non-Faradic charge injections are used to obtain a high energy density in an asymmetric supercapacitor, and the results presented here indicate that adjusting electrode initial potential is a simple but effective strategy to enhance the energy density of asymmetric supercapacitors.

2. Experimental

2.1. Synthesis of porous V₂O₅ nanotubes

Porous V₂O₅ nanotubes were prepared by electrospinning. In a typical synthesis, 1.25 g ammonium vanadate (NH₄VO₃), 2.72 g oxalic acid dihydrate (H₂C₂O₄·2H₂O), and 1.1 g polyvinylpyrrolidone (PVP, Mw = 1,300,000) were dissolved in a hybrid solution of 10 ml absolute ethanol (C₂H₅OH) and 10 ml deionized water under vigorous stirring for 12 h to form a homogeneous precursor solution. Then the viscous mixture was loaded into a plastic syringe equipped with a 7 # stainless steel needle. Direct current voltage of 15 kV was applied between needle tip and the aluminum foil collector with a distance of 15 cm, at a flow rate 1 ml h^{–1}. Subsequently, the electrospun nanofibers collected from aluminum foil were calcined at 400 °C for 15 min in a muffle furnace at a heating rate of 0.5 °C min^{–1}.

2.2. Synthesis of activated carbon nanorods

Activated carbon nanorods were prepared by carbonization of polyaniline (PANi) with KOH activation. The detailed synthesis method of activated carbon nanorods could be found in our previous work [31,32].

2.3. Characterization

The crystallographic structures of porous V₂O₅ nanotubes was determined by power XRD system (XRD, Rigaku D/Max-2400) with Cu K α radiation ($\lambda = 1.5418 \text{ \AA}$). The morphology and microstructure of electrode materials were characterized by field emission scanning electron microscopy (FESEM, Hitachi S4800) and transmission electron microscopy (TEM, JEOL 2100 FEG). The specific surface area and pore structure of activated carbon nanorods were analyzed by Brunauer-Emmett-Teller (BET) method.

2.4. Electrochemical characterization

The working electrodes were prepared by mixing electroactive materials, acetylene black conducting graphite and poly (tetrafluoroethylene) in a mass ratio of 80:7.5:7.5:5 with a few drops of absolute ethanol to obtain homogeneous slurry. Then the slurry of V₂O₅ was pressed onto carbon cloth current collector (1 cm²) as positive electrode, and the slurry of activated carbon nanorods was pressed onto nickel foam current collector (1 cm²) as negative electrode and dried at 60 °C for 12 h in air. Electrochemical measurements of individual working electrode were performed in a three-electrode system by using an electrochemical workstation (CHI660D, Shanghai, China), in which a platinum foil electrode and a saturated calomel electrode were used as the counter electrode and the reference electrode in 2 M LiNO₃ aqueous electrolyte. The working potential windows are –0.2 to 0.8 V for V₂O₅ nanotubes electrode and –0.9 to 0 V for carbon nanorods electrode, respectively. Electrochemical impedance spectra are recorded from 100 kHz to 0.01 Hz. The specific capacitances (C_m) were calculated from galvanostatic discharge curves using the equation $C_m = (I \cdot \Delta t) / (m \cdot \Delta E)$. Where I is discharge current, Δt is discharge time, m is the mass of electro-active material and ΔE is the discharge voltage range. In an asymmetric supercapacitor system, the m meant the total mass of positive and negative electro-active materials. And the average specific capacitances (C_s) could also be calculated by the equation $C_s = S / (2m \cdot v \cdot \Delta E)$ according to the CV curves. Where S is area of CV curves, v is sweep rate. To construct the V₂O₅//C asymmetric supercapacitor, the mass ratio of positive and negative electrodes was 1:1 to obtain charge balance between

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