



# High-performance aqueous asymmetric supercapacitor based on carbon nanofibers network and tungsten trioxide nanorod bundles electrodes



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## ABSTRACT

The demand for high-performance energy storage devices such as supercapacitors and lithium-ion batteries has been increasing to meet the application requirements of renewable energy systems. Here, high energy density aqueous asymmetric supercapacitor (ASC) is assembled based on carbon nanofibers (CNF) network positive electrode and tungsten trioxide (WO<sub>3</sub>) nanorod bundles negative electrode. Polyaniline-based CNF are prepared by direct carbonization of polyaniline nanofibers. WO<sub>3</sub> nanorod bundles are synthesized via a simple sodium chloride assisted hydrothermal process. The CNF//WO<sub>3</sub> ASC device operates with a voltage of 1.6 V and achieved a high energy density of 35.3 Wh kg<sup>-1</sup> at a power density of 314 W kg<sup>-1</sup>. Furthermore, the device shows an excellent cycling performance with capacitance retention of 88% after 1000 cycles.

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## 1. Introduction

Supercapacitors are a class of energy storage devices which have a much higher power density and longer cycling stability than conventional batteries [1,2]. However, their energy density is still relatively low compared with batteries and fuel cells. Therefore, one of the most critical aspects in the development of supercapacitors is to enhance their energy density without sacrificing high power capability [3]. The energy density ( $E$ ) of the supercapacitor can be enhanced by either increasing the device capacitance ( $C$ ) or broadening the cell voltage ( $V$ ) according to the equation  $E = 0.5CV^2$ . In generally, organic electrolytes used in supercapacitors can provide a more wide electrochemical window. However, organic electrolytes have poor electrical conductivity and environmentally unfriendly [4]. Conversely, aqueous electrolytes have significant inherent advantages over organic electrolytes including low-cost, high ionic conductivity, inherently safe, and so on [5]. Besides having a wide electrochemical window, to develop a high energy density supercapacitor device, an active electrode material with high capacity performance is indispensable. In addition, asymmetric supercapacitors (ASCs) consisting of two different

electrodes are designed to promote the specific energy in comparison to symmetric supercapacitors. This is because ASCs have a wider operating voltage window (more than 1.5 V) due to kinetic suppression of oxygen evolution on the electrode surface [6].

The challenge of ASCs is how to make matching between the positive electrode and negative electrode in a suitable electrolyte to obtain the highest acceptable cell voltage without compromising energy density, power density, and cycle-life [7]. Most of the ASCs are developed using metal oxide as positive electrode because the majority of the metal oxides work at positive potential window [8]. Meanwhile, most of researcher use carbon based materials as negative electrode. Unfortunately, the energy density of electrode assembly cannot be increased substantially due to their low device capacitance or narrow useful match voltage window [5,9–10]. In addition, it is well known that the low electronic conductivity of most metal oxides profoundly affects their electrochemical performance. In order to solve this problem, the use of integrated metal oxide-based electrode [11] or some high-conductivity nanomaterial, such as tungsten oxide nanowires [12], as an advanced electrode materials is proposed. Furthermore, it is found that some metal oxides such as MoO<sub>3</sub>, WO<sub>3</sub> and Bi<sub>2</sub>O<sub>3</sub> can be used as negative electrode since they involve in the redox reaction within the negative potential [13–15]. Meanwhile, carbon based materials can be used as a positive electrode for ASC due to it can works work at either positive and negative potential and there is no chemical reaction occurred in

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between the electrolyte and electrode. Hence, the novel ASCs are developed using nanostructured metal oxides as negative electrode and carbon based materials as positive electrode has aroused wide concern [15,16]. Typically, Tang et al. prepared aqueous asymmetric supercapacitor using  $\text{MoO}_3$  nanoplates as an anode material, activated carbon as cathode material, which ASC can deliver a high energy density and good rate behavior [16]. Although ASCs based on metal oxide and carbon based electrode materials were successfully assembled, so far few studies reported the fabrication of ASCs with carbon based materials as positive electrode and  $\text{WO}_3$  as negative electrode in aqueous electrolytes. Nowadays, carbon based materials, especially 3D carbon nanostructures (such as carbon nanofiber network), have extensively used as electrode materials for advanced supercapacitor applications in view of their low cost, hierarchical porous channels, high electrical conductivity and better cycling stability [17,18]. Similarly, tungsten trioxide is an n-type semiconductor with high electrical conductivity [19] and different crystal structures that provide a suitable structure for intercalation of small cations like  $\text{H}^+$  [20].

In the present work, we focused on enhancing the energy density and long cycling stability of ASC based on polyaniline-based carbon nanofibers (CNF) network positive electrode and tungsten trioxide ( $\text{WO}_3$ ) nanorod bundles negative electrode. In 1 M  $\text{H}_2\text{SO}_4$  aqueous electrolyte, the as-made CNF/ $\text{WO}_3$  asymmetric supercapacitor exhibits superior electrochemical capacitive properties, including wider operating voltage window (1.6 V), high energy density of  $35.3 \text{ Wh kg}^{-1}$  at a power density of  $314 \text{ W kg}^{-1}$  and outstanding cycle stability.

## 2. Experimental

### 2.1. Materials and chemicals

Aniline monomer (AN, Aladdin Ltd. Shanghai China) was distilled under reduced pressure. Ammonium persulfate (APS, Aladdin Ltd. Shanghai China), sodium alginate (Aladdin Ltd. Shanghai China),

sodium chloride (NaCl, Tianjing Chemical Co., China) and sodium tungstate dihydrate ( $\text{Na}_2\text{WO}_4 \cdot 2\text{H}_2\text{O}$ , Aladdin Ltd. Shanghai China), all chemical reagents were in analytical grade.

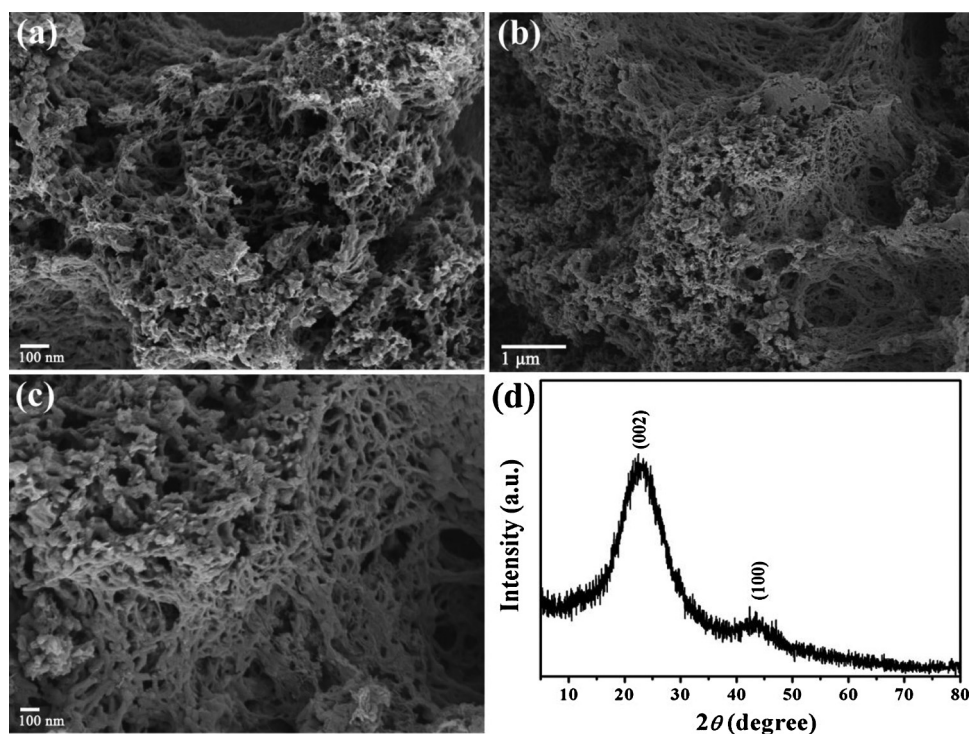
### 2.2. Synthesis of polyaniline nanofibers and polyaniline-based carbon nanofibers network

The polyaniline nanofibers were prepared similar to the literature [21]. In a typical process, 0.05 g sodium alginate was dissolved in 70 mL 1 M HCl solution at ambient temperature, and then 2.0 g aniline monomer was introduced into the above solution. After being stirred for 30 min, 5.75 g ammonium persulfate was dissolved in 10 mL deionized water and added into the above system. The polymerization was performed for 12 h at room temperature. Finally, the product was collected by centrifugation and washed successively with deionized water and ethanol until the filtrate was colorless, and then dried at  $60^\circ\text{C}$  for 24 h to obtain a dark green powder.

The as-synthesized PANI nanofibers were carbonized under high pure  $\text{N}_2$  atmosphere at  $800^\circ\text{C}$  with a heating rate of  $5^\circ\text{C min}^{-1}$ , and maintained for 2 h at this temperature to form nitrogen-doped carbon nanofibers network. The obtained product was denoted as CNF.

### 2.3. Synthesis of $\text{WO}_3$ nanorod bundles

$\text{WO}_3$  nanorod bundles were synthesized by a sodium chloride assisted hydrothermal method. Briefly, 2.0 g sodium tungstate dihydrate and 0.5 g sodium chloride was dissolved in 60 mL deionized water, and a 3 M HCl aqueous solution was used to adjust the pH value to 2.0. After stirring for 2 h, the solution obtained was transferred into a 100 mL Teflonlined stainless autoclave and heated at  $180^\circ\text{C}$  for 24 h. After cooled to room temperature naturally, the resulting precipitates were collected by filtration, washed with distilled water and absolute ethanol for several times to remove the residue of reactants, and then dried in vacuum at  $60^\circ\text{C}$  for 12 h.



**Fig. 1.** (a) SEM images of as-synthesized polyaniline nanofibers; (b and c) SEM images of CNF at different magnification; (d) XRD pattern of CNF.

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