



## Regular Article

## All-round utilization of biomass derived all-solid-state asymmetric carbon-based supercapacitor

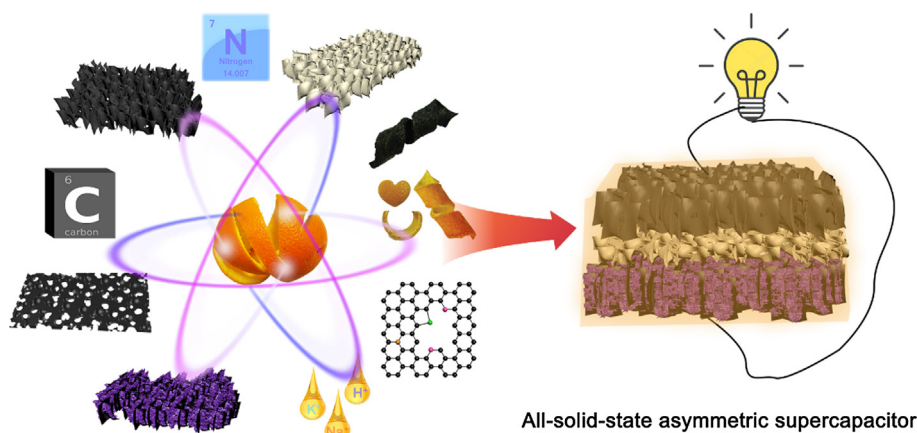
Chao Wang, Ye Xiong, Hanwei Wang, Qingfeng Sun\*

School of Engineering, Zhejiang A&amp;F University, Hangzhou 311300, China



## GRAPHICAL ABSTRACT

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## ARTICLE INFO

## Article history:

Received 18 March 2018  
 Revised 20 May 2018  
 Accepted 29 May 2018  
 Available online 30 May 2018

## Keywords:

Biomass  
 All-round utilization  
 Carbon  
 All-solid-state  
 Supercapacitors

## ABSTRACT

All-round utilization of resources is proposed for maximizing environmental and economic benefits. Herein, the concept of all-round utilization on biomass derivations applying to carbon-based supercapacitors is demonstrated. Orange peel is used for all subassemblies of supercapacitor, including electrodes, separator and electrolyte. A monolithic porous carbon (OPHPC) is prepared by one-step carbonization of orange peel and another composite electrode is further synthesized by a simple hydrothermal process, based on sufficient utilization of natural structure and chemical components. OPHPC exhibits a high specific surface area of  $860 \text{ m}^2 \text{ g}^{-1}$  and naturally doped nitrogen. The composite electrode shows the homogeneous and high mass loading of  $\text{MnO}_2$ . Orange peel also affords the role of separator benefited from the natural porous channel structure and high porosity of 74.6%. Orange peel juice is exploited to produce the electrolyte, and exhibits the best retention in natural separator. All-orange peel all-solid-state supercapacitor shows the high areal capacitance of  $3987 \text{ mF cm}^{-2}$ . Furthermore, the flexibility of orange peel is also utilized to achieve the shape-tailored monolithic porous carbon electrode and device, which further extends the utilized dimensionality in biomass applying to supercapacitors. The work starts with all dimensional utilization for biomass derived supercapacitor.

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\* Corresponding author.

E-mail address: [qfsun@zafu.edu.cn](mailto:qfsun@zafu.edu.cn) (Q. Sun).

## 1. Introduction

Excessive exploitation of fossil fuels has caused the environmental and socioeconomic concerns associated with their use [1,2]. The rapid society developing required plenty of the available resources, whereby a paradigm shift was to displace conventional fuel with renewable resources [3,4]. Biomass was a nearly limitless renewable resources from the production of photosynthesis [5,6]. Sufficient utilization of biomass resources is hopeful for further energy security and global carbon balance. Among, biomass-derived carbon-based supercapacitor was an attractive approach in the current years [7–13]. But, in a strict sense most work could only be defined as biomass-derived porous carbon powders for supercapacitor electrodes. So the biomass precursor only played a role of donator of carbon element, that is, single utilization mode. All-round utilization of resources was desiring because it based on comprehensive consideration for the environmental and economic benefits [14,15]. Importantly, biomass materials congenitally possess many properties [16], and thus they could be multiply used.

Natural organisms evolving over millions of years always possessed the unique structure with geometry configuration [17]. The efficient architecture engineering was able to promote electrochemical performance of electrode materials [18,19]. So the unique natural structure of biomass materials should be fully considered and utilized for derived carbonaceous materials. On the other hand, the chemical components, except for carbon element, of biomass precursor needed to be given crucial attention, because the heteroatoms (mainly concluding O, N, S, P, and B) could efficiently improve the electrode performance through increasing the surface Faradaic reaction [19]. Furthermore, other some natural compositions (such as biotic acids or some salts) also should be effectively applied for the activation (forming the porous), but they often was neglected due to, in most cases, the existence of external activation agent. Whereby, the appropriate biomass precursors could realize carbonization and high porosity at the same time under high-temperature treatment by self-activation [20,21]. This process without chemical activation agent was more environmental friendly and contributed to the retention of beneficial heteroatoms. Besides, the natural organisms contain large amounts of the aqueous solution with abundant ions, which could be used as a natural source of aqueous electrolyte. It not only could extend a dimension of utilization of biomass in the derived supercapacitors, but also met the requirement for maximum utilization of the resource.

Supercapacitors had attracted great attention in a huge demand because of their eminent features, for instance, the prodigious cycle life, fast recharge capability, low maintenance, and high power density [22,23]. Choices of all-solid-state and asymmetric-type electrodes were advisable for supercapacitors devices in practical application due to its high capability and energy density without sacrifice of cycle life [24–26]. Meanwhile, in the practical application, electrodes with high thickness/load mass were required because of the effective components and packing density of devices, which could bring about the lower expending [27,28]. But, the ion and electron transport usually had difficulty transport in the thickness electrodes, especially yet absence of directional pore channel. As a result, low tortuosity thick monolithic wood AC was reported to address the challenge [28]. Nevertheless, the designated tortuosity or shape probably was in demand for diverse practical application of supercapacitors (such as some corner or narrow space inside the device).

Hence, in this works, not only porous monolithic carbon electrode was synthesized by one-step annealing process considering the utilization of natural structure and components of orange peel, but also using its plasticity/flexibility and inherent juice fabricated the ultimate shape-tailored all-solid-state hybrid supercapacitors. Hierarchical porous characteristics of OPHPC benefited from inher-

itance of natural structure and self-activation. It not only possessed well capacitive performance but also the unique pore channel structure could effectively load inorganic nanomaterials. The electrolyte directly came from the squeeze juice of orange peel, and the shape-tailored feature also utilized flexibility of orange peel. This work firstly achieved the all-dimensional utilization of biomass for fabricating the all-solid-state asymmetric supercapacitors. The ultimate all-orange peel derived and shape-tailored all-solid-state asymmetric supercapacitors showed a leading areal capacitance.

## 2. Experimental section

### 2.1. The synthesis of OPHPC and MnO<sub>2</sub>@OPHPC

Orange, *citrus sinensis*, purchase from the fruit supermarket, and then the orange peel was obtained. Next, orange peel was cut into the appropriate size and thickness (using the epicarb). Subsequently, the orange peel was placed in a horizontal tube furnace under the protection of nitrogen and heating rate was set as 5 °C min<sup>-1</sup> by the following procedure: it was firstly heated to 150 °C and was kept at this temperature for 60 min to further remove the water; then the temperature was increased to 800 °C and hold for 3 h. The obtained samples were donated by the OPHPC materials and directly were used as the electrode materials (as anode for the asymmetric supercapacitors). In addition, orange peel could be defined into diverse shapes by the mold tools. And, the bending and even spiral orange peel underwent the simple drying to finalize the shapes.

The MnO<sub>2</sub>@OPHPC composite (as cathode of asymmetric supercapacitors) was prepared by the hydrothermal process. Firstly, 1.58 g of KMnO<sub>4</sub> was dissolved in 50 mL of the deionized water, forming a solution. Afterwards, the OPHPC materials were fully immersed into the prepared KMnO<sub>4</sub> solution by stirring with a magnetic bar at room temperature for 10 h. Thereafter, the precursor was transferred into a teflon-lined stainless steel autoclave, sealed and kept in an oven at 180 °C for 60 min, then naturally cooled down to room temperature. The obtained samples were washed by the deionized water several times and were then dried at 80 °C for 24 h. For the other metal oxides and metal sulfides (including ZnO, Fe<sub>3</sub>O<sub>4</sub>, TiO<sub>2</sub>, and MoS<sub>2</sub>), the corresponding composites were fabricated by hydrothermal treatment or the vacuum-assisted infiltration, also demonstrated in Table S1.

### 2.2. Characterizations

Field emission SEM (Quanta FEG 250) and TEM (Tecnai G2 F20, USA) were applied to look into the microstructure of these samples. X-ray powder diffraction was conducted using a Bruker D-8 Advance diffractometer (USA) with nickel-filtered Cu K $\alpha$  ( $\lambda = 1.54178 \text{ \AA}$ ) radiation at room temperature and the data was collected from 5° to 80° with a scan rate of 2° min<sup>-1</sup>. XPS was conducted with two separate systems equipped with monochromatic Al K $\alpha$  sources (ThermoFisher K-Alpha XPS system, USA) to analyze the chemical composition of the samples. Ca, K, Mg, Na, Fe, Zn and Cu elements was determined by Inductively Coupled Plasma (ICP, VISTA-MPX, USA). Raman spectrum was recorded with 532 nm excitation using a multi-wavelength micro-Raman spectroscope (Renishaw RM2000). The measurements of the N<sub>2</sub> adsorption isotherms were performed with Micromeritics ASAP 2020 surface area and pore size analyzer (Micromeritics instrument Ltd., USA).

### 2.3. Electrochemical performances

All electrochemical performances were evaluated by using a CHI 660 E electrochemical workstation (Shanghai Chenhua

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