

# Porous carbons produced by the pyrolysis of green onion leaves and their capacitive behavior

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**Abstract:** Porous carbons were prepared by the simple carbonization of green onion leaves at temperatures from 600 to 800 °C and used as the electrode materials of supercapacitors. SEM, FESEM, EDX, AAS, XRD, TGA and nitrogen adsorption were used to characterize their morphology, pore structure and surface elemental composition. Cyclic voltammetry, electrochemical impedance spectroscopy and galvanostatic charge/discharge were carried out to evaluate their specific capacitance, resistance and cycling life. Results showed that the initial mineral elements present in the leaves such as calcium (Ca) and potassium (K) play an activating role during the carbonization. All samples have a bimodal pore distribution of micropores (mainly 0.6-1.2 nm) and mesopores (mainly 3-5 nm). The carbon prepared at 800 °C had the highest surface area of 551.7 m<sup>2</sup>/g, a specific capacitance of 158.6 F/g at 0.2 A/g and an effective areal capacitance of 28.8 μF/cm<sup>2</sup>. The effective areal capacitance of the carbon prepared at 800 °C is higher than of most porous carbons reported in the literature, which is ascribed to its pore size distribution that favors ion access to its pores.

**Key Words:** Porous carbon; Green onion leaves; One-step carbonization and activation; Effective areal capacitance

## 1 Introduction

With the increase of the environmental pollution and the scarcity of fossil fuels, the demand for clean energy sources is growing rapidly all around the world. Supercapacitor, as a kind of clean energy conversion and storage device, has attracted much attention owing to its high power density, long cycle life and high dynamic of charge propagation, which bridges the power/energy gap between traditional dielectric capacitor and battery<sup>[1-6]</sup>. Especially, electrical double-layer supercapacitors (EDLs), draw much more attention owing to their simple charging mechanism, long cycling life and short charging time. Since pure physical charge accumulation occurs at the electrochemical interface between electrode and electrolyte during the charge/discharge process, EDLs are able to store and deliver energy at a relatively high rate<sup>[7-10]</sup>. Compared to batteries, supercapacitors have the advantages of high power density, long life expectancy, long shelf life, high efficiency, wide range of operating temperatures, environmental friendliness and safety. However, they also face challenges at the current stage of technology, such as low energy density, high cost and high self-discharging rate. Among the components of a supercapacitor, electrode materials dominate the performance of supercapacitors<sup>[11]</sup>. Therefore, developing new materials with improved performance is important to improve the property of supercapacitors<sup>[12]</sup>. In general,

electrode materials of supercapacitors include three types<sup>[13,14]</sup>: carbon materials, conducting polymers, and metal oxides. Porous carbons have large surface areas, relatively good electrical conducting properties and the 3D porous network structure that ensures fast electronic and ionic conduction through charge/discharge process. Furthermore, porous carbons are considered as the most promising candidate materials for supercapacitors in industry owing to their moderate cost<sup>[3,7,15]</sup>. Generally, the synthesis of porous carbons includes two steps: carbonization and activation. Among various precursors, cheap and renewable biomass such as agricultural byproducts have attracted much attention owing to their low cost and environmental friendly properties<sup>[16-18]</sup>. Activation is a crucial procedure, which include physical and chemical activation. For these two methods, either high temperature or large amount of chemical agent is used, which require expensive equipments or bring about difficulty in post-treatment<sup>[19-25]</sup>. Though various porous carbons have been tried as electrode materials in supercapacitors, their applications are still limited owing to their complicated production processes<sup>[26]</sup>. As reported, natural constituents such as mineral substances in some kinds of leaves may replace the additional pore generators to create micropores, thereby simplifying the process<sup>[27,28]</sup>. Green onions are widely planted in China and could be stored in winter. However,

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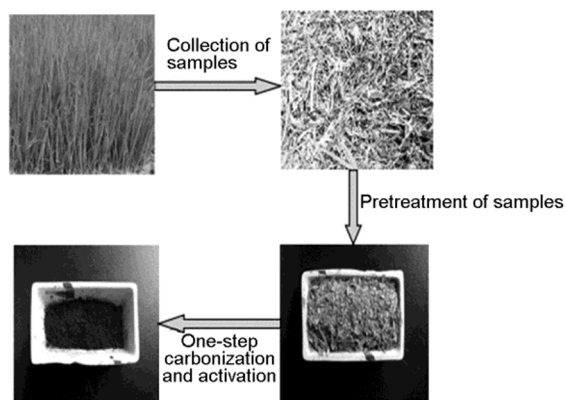


Fig. 1 Schematic diagram for the synthesis of porous carbons from green onion leaves.

during the storage, the leaves of green onions are usually withered and need to be discarded. Therefore, we reported a facile, cost-effective approach to synthesize porous carbon via one-step pyrolysis of the discarded green onion leaves without any additive. The reason might be that green onion leaves contain Ca and K that act as pore generators<sup>[27,28]</sup>. The pore sizes are mainly centered around 0.6-1.2 and 3-5 nm. Although the specific surface area and the mass specific capacitance for the green onion leaf-derived carbons (GOLCs) are not so high, their “effective areal capacitance” is high, indicating that the proportion of their effective pores in GOLCs is high.

## 2 Experimental

### 2.1 Chemicals

The green onions used in this study were directly obtained from the local farm. Nafion solution was purchased from Sigma. All other chemical reagents, such as hydrochloric acid (HCl, 36%), nitric acid (HNO<sub>3</sub>, 65%), perchloric acid (HClO<sub>4</sub>, 70%), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>, 30%) and potassium hydroxide (KOH, 98%), were purchased from Sinopharm Chemical Reagent Co. Ltd and used as received without any further purification. All the aqueous solutions were prepared with Millipore water having a resistivity of 18.2 MΩ (Purelab Classic Corp., USA).

### 2.2 Synthesis of porous carbons

The synthesis process of green onion leaf-derived carbons (GOLCs) is shown in Fig. 1.

The leaves of green onion were separated from the white stem, washed thoroughly with deionized water and dried at 60 °C in an oven over night. The dried leaves were crushed into powder. The carbonization and activation processes were carried out at one step. The dried leaf powder was heated at 600-800 °C under the protection of argon for 2 h in a tubular furnace. The heating rate was 10 °C/min. After cooled down to room temperature under argon, the green powder was totally

turned into black color. The obtained products were washed thoroughly by deionized water and then dried in an oven over night. For comparison, some products were rinsed by a diluted hydrochloric solution (0.1 M).

### 2.3 Electrochemical measurements

Electrochemical characterizations were carried out in a three-electrode electrochemical system using Hg/HgO electrode and platinum foil as the reference and counter electrode, respectively. The GOLC powder was dispersed in water by sonication. Then the suspension was dripped on a glassy carbon electrode and coated by Nafion solution.

All the electrochemical measurements were carried out on a CHI 660D electrochemical workstation (Shanghai Chenhua Co. Ltd, China) in 3M KOH aqueous electrolyte solution at room temperature. Cyclic voltammetry (CV) curves were obtained between a potential range of -1.0- 0.1V at different scanning rates. The electrochemical impedance spectroscopy (EIS) was performed in a three-electrode system at 5 mV-alternating current-disturbance around the open circuit potential vs Hg/HgO. The scanning frequency was from 0.01 to 100 kHz. The galvanostatic charge/discharges (GCD) were carried out under different current densities.

The mass specific capacitance is calculated from GCD curves through equation (1) :

$$C_s = \frac{2I}{m} \frac{dt}{dv} \quad (1)$$

Where “C<sub>s</sub>” is the specific capacitance, “I” is the current, “m” is the active mass and “dv/dt” is the slope obtained from the discharge curve.

Effective areal capacitance (C<sub>ea</sub>, μF/cm<sup>2</sup>) means the ratio of “mass specific capacitance (C<sub>ms</sub>, F/g)” and “BET surface area (A, m<sup>2</sup>/g)”, which is calculated by the equation (2).

$$C_{ea} = \frac{C_{ms}}{A} \quad (2)$$

### 2.4 Characterizations

The morphology of GOLCs was observed by a JSM-6510LV (Japan) scanning electron microscope (SEM) and a JSM-7800F field-emission scanning electron microscope (FE-SEM, Japan). Elemental composition analysis was qualitatively measured by JSM-6510LV (Japan) energy dispersive X-ray spectroscopy (EDX) and quantitatively determined by WFX-110 flame atomic absorption spectrometry (FAAS). The samples were pretreated before FAAS measurement. Firstly, they were ground into powder and poured into an acid mixture of HNO<sub>3</sub> and HClO<sub>4</sub>, followed by heating and dissolving at a hot plate until most of water evaporated. Then H<sub>2</sub>O<sub>2</sub> was added to get rid of the residual acid. Through the treatment, minerals such as K and Ca could be totally

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