Ulothrix-Derived Sulfur-Doped Porous Carbon for High-Performance Symmetric Supercapacitors
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ABSTRACT: With the demand for carbon dioxide emission reduction, the sustainable conversion of useless biomass into high-value energy storage devices has received excellent scientific and technological attention. The high synthesis cost and low specific capacitance limited the supercapacitor application. Therefore, biomass-derived sulfur-doping porous carbon (SPC) has been synthesized from ulothrix using simple pyrolysis and chemical activation methods. The unique activated carbon material exhibits a high specific surface area (2490 m² g⁻¹), and the effect of the activator addition ratio was systematically investigated. The optimized SPC-2 displayed a high specific capacitance (324 F g⁻¹ at 1 A g⁻¹) and excellent cycling stability (90.6% retention after 50 000 cycles). Furthermore, a symmetric supercapacitor (SSC) based on SPC-2 demonstrated a high energy density (12.9 Wh kg⁻¹) at an 800 W kg⁻¹ power density. This work offers a simple, economical, and ecofriendly synthetic strategy of converting widespread, useless biomass waste into high-performance supercapacitor applications.

1. INTRODUCTION
With the increasing demand for portable power storage systems in automobiles, electronic goods, and capital machinery, researchers have expended a great amount of effort to develop electric energy storage devices with high energy density and output power. As a novelty electric device filling the gap between batteries and traditional capacitors, supercapacitors have satisfactory energy densities and power densities.¹ The performance of supercapacitors depends mainly on the electrode materials, and a variety of novel nanostructured carbon materials have been widely adopted.² They can be classified into electrical double-layer capacitors (EDLCs)³ according to the charge-storage mechanism, and another type consists of pseudocapacitors. For these electrical double-layer carbon materials, a large surface, suitable pore structure, and high conductivity are essential to storing electric charge.

To meet these requirements, researchers mainly consider the improvement from the aspects of proper carbon sources,⁴ the activation procedure,⁵ and heteroatom doping.⁶ Many kinds of precursors were selected as carbon sources to boost the surface properties, such as biomass-derived carbon,⁷⁻¹⁰ polymer-derived carbon,¹¹,¹² carbide-derived carbon,¹³ and MOF-templated carbon.¹⁴¹⁵ Among these, biomass has been taken into account by many researchers because of its large specific area, abundant pore structure, ecofriendliness, and extensive sources.¹⁶ For example, various biomass products such as walnut shells,¹⁷¹⁸ pomelo peels,¹⁹ bamboo bagasse,²⁰ willow catkins,²¹ cornstalks,²² bean curds,²³ and garlic seed²⁴ were made into porous carbon. Woody biomass mainly consists of cellulose, hemicellulose, and lignin.²⁵

Activation is a crucial process for pore formation in biomass materials.²⁶ A large amount of carbon exists in the heating process and maintains its original morphology, which is the best natural template.²⁷²⁸ Meanwhile, gases escape, forming channels. After chemical activation, biomass becomes porous carbon with large specific surface areas and develops a pore structure.

Besides, biomass carbon has the advantage of self-doping because these natural products have many organic substances. The tissues can convert to heteroatom doping in the carbon matrix during pyrolysis, avoiding extra reagents. And these heteroatoms, such as nitrogen,²⁹ oxygen, sulfur,³⁰ and phosphorus,³¹ can efficiently enhance electrical conductivity and wettability.

Ulothrix is widely grown in most parts of our planet, and its excessive growth is often regarded as a sign of water body eutrophication. The government pays the bill to clean it up...
every year, whereas it has potential resource utilization value. As shown in Scheme 1, ulothrix is green and filiform, serving as the natural template for carbon microbelts. The floristic stoma, along with the abundant pores formed in the carbonization and activation process, results in rich hierarchical apertures. In addition, fibrin and amino acids in the plant tissue can be considered to be sulfur and nitrogen species to obtain self-doping carbon materials. These doped atoms can efficiently enhance the conductivity of carbon skeletons, which is helpful for electrochemical performance.

Herein, a sulfur-doped porous carbon with a large specific surface area is obtained by pyrolysis and following chemical
activation. The unique characterizations of the SPC are the following: (i) ulothrix with a proper microbelt shape and rich organic matter is selected as the natural template; (ii) the synthetic process is simple and easy without the use of extra templates and a postdoping procedure; and (iii) the carbon product has a large surface area and uniform sulfur doping. Such a low-cost, ecofriendly, high-performance carbon converted from useless plant tissue to a high-value product is economically beneficial and highly feasible.

2. RESULTS AND DISCUSSION

2.1. Characterization of Materials. An illustration of the synthesis of the SPC is shown in Scheme 1. By cleanly heating ulothrix in an inert atmosphere at 500 °C for 1 h, the pyrolytic carbon powder was prepared. Then the pyrolytic carbon mixed with KOH was heated to 800 °C for 2 h. Finally, the SPC was acquired by using excess hydrochloric acid, washing the products until the pH was below 7, and drying in vacuum at 80 °C for 12 h. The XRD patterns of the SPC samples are shown in Figure 1a, and all three curves show two typical diffraction peaks at 24 and 43°, corresponding to the (002) and (100) planes of the graphite phase (PDF no. 89-8487). These weak, broad peaks indicate that after pyrolysis and activation the SPC planes of the graphite phase (PDF no. 89-8487). These weak, broad peaks indicate that after pyrolysis and activation the SPC is considered to be amorphous carbon with a low degree of graphitization.32 The Raman spectra (shown in Figure 1b) of the SPC samples exhibit two base peaks at 1340 and 1595 cm−1, which correspond to the disordered graphite carbon phase (D band) and the graphic carbon phase (G band), respectively. The intensity ratios of the D/G band for SPC-1, SPC-2, and SPC-3 are 0.929, 0.963, and 0.974, indicating the imperfect structure of the graphitic carbon.34 The morphology of the samples was explored by using the SEM (Figure 2). It is clear that the clean ulothrix (Figure 2a,b) displays a belt shape that is about 40–80 μm wide and a few micrometers thick. After pyrolysis (Figure 2c,d), the particles displayed many wrinkles on a smooth surface as the plant tissues lost water. By comparison, SEM images of the activated SPC samples are shown in Figure 2e–j. All SPC samples indicate many small irregular particles with hierarchical pore structure, and various sizes of holes and channels can be found. It is clear that mesopores of a few nanometer sizes can be found in the high-resolution image of SPC-1 (Figure 2f), resulting from the high-temperature activation process. For SPC-2 and SPC-3, there were more pores on the surface. TEM images further display the mesoporous structure of the carbon sheets (Figure 2k,l). The specific surface areas, pore size distributions, and pore volumes of the SPC samples are calculated on the basis of the N2 adsorption test results (Table 1). The specific surface areas of SPC-1–SPC-3 are 1944, 2490, and 2018 m2 g−1, and the pore size distributions of SPC-1–SPC-3 are located at 4.32, 5.33, and 6.10 nm, respectively, revealing the mesoporous structure in the carbon sheets.

Table 1. Specific Surface Areas, Pore Size Distributions, and Pore Volumes of SPC

| sample | surface area (m2 g−1) | pore volume (cc g−1) | pore diameter (nm) |
|--------|-----------------------|----------------------|--------------------|
| SPC-1  | 1944                  | 0.615                | 4.32               |
| SPC-2  | 2018                  | 0.532                | 5.33               |
| SPC-3  | 2490                  | 0.835                | 6.10               |

Nitrogen adsorption/desorption isotherms and pore size distribution diagrams are shown in Figure S1. To verify the elemental makeup of the samples, XPS measurements were conducted, and the spectra of SPC-2 are shown in Figure 3. The C, O, and S element contents (atom %) are 79.74, 16.46, and 2.2%, respectively. The full spectrum shows obvious O 1s, C 1s, and S 2p peaks. The C 1s narrow peak can be indexed to the fitting peaks at 288.64, 285.96, and 284.67 eV, and the S 1s peak can be fitted to three peaks: S1 at 163.8, S2 at 164.9, and S3 at 168.7 eV. The S1 and S2 peaks correspond to S2p3/2 and S 2p1/2 of the C–S–C covalent bond, and the S3 peak can be indexed to the oxidized sulfur C=S–O=C bond following previous reports.30,35,36 For the O 1s spectrum, the O1 peak at 531.87 eV can be attributed to the carbon–oxygen bond (C=O). The O2 peak at 533.23 eV corresponds to the binding energies of oxygenated C–S functionalities (C=S–O).25 These results demonstrate that sulfur atoms are successfully incorporated into the carbon network and play an essential role in enhancing the electrochemical performance of SPC.

2.2. Electrochemical Performance of the SPC Electrodes. The electrochemical performance of SPC electrodes was first evaluated by CV measurement. As shown in Figure 4a, all of the CV curves at a scan rate of 100 mV s−1 have a rectangular shape, indicating typical electric double-layer capacitor behavior. The CV curves at different scan rates of SPC-1 and SPC-3 are shown in Figure S2. Among them, SPC-2 has the highest oxidation peak and the largest enclosed area. The GCD measurement resulting in a current density of 1 A g−1 are performed in Figure 4b, and all of the GCD curves show a symmetric triangle shape. From the discharge time, the electrochemical performance of SPC. The GCD curves at different current densities of SPC-1 and SPC-3 are shown in Figure S3. On the basis of the GCD measurement result, the corresponding specific capacitances of the SPC electrodes at different current densities are shown in Figure 4c. At 0.5 A g−1, the SPC-1–SPC-3 electrodes display the highest specific
capacitances of 116.31, 390.38, and 327.50 F g\(^{-1}\). Even at 20 A g\(^{-1}\), the specific capacitances were 40.71, 212.46, and 110.29 F g\(^{-1}\). EIS measurements were performed to estimate the impedance of the SPC electrode (Figure 4d). All of the EIS curves consisted of a steep slope in the low-frequency zone and a depressed semicircle in the high-frequency zone. The charge-transfer resistance (\(R_{ct}\)) was fitted by Zview software based on the equivalent circuit inserted in Figure 4d.\(^{37}\) The charge-transfer resistance (\(R_{ct}\)) values of the SPC electrode were 0.21, 0.22, and 0.84 \(\Omega\). The intrinsic resistance (\(R_s\)) values of the SPC electrode were 0.91, 0.81, and 1.05 \(\Omega\). It is clear that SPC-2 has the lowest inherent resistance.

Cycling performance tests were conducted to evaluate the lifespan of the SPC electrodes. As shown in Figure 4e, in 10 000 charge/discharge cycles, the SPC electrodes showed very little decay in electrochemical capacity. After the long cycles, the SPC-1–SPC-3 electrode still retain 88.2, 210.2, and 129.6 F g\(^{-1}\), and the corresponding capacity retentions are 92.0, 91.0, and 87.8%.

Because the SPC-2 electrode exhibited the highest capacity of the studied electrode, it was further estimated at different scan rates (Figure 5a). The oxidation and reduction peak current increased with increasing scan rate. Meanwhile, the curves retain a rectangular shape, suggesting electric double-layer behavior. GCD tests were also performed at different current densities to evaluate the rate performance of SPC-2. As shown in Figure 5b, at current densities of 1, 2, 4, 8, 10, and 20 A g\(^{-1}\), it delivers 325.51, 247.57, 221.04, 213.89, 212.50, and 212.46 F g\(^{-1}\), respectively. The SPC-2 electrode has 73.9%
When the voltage surpasses 1.4 V, the current oxidation increases sharply, implying oxygen evolution. For the 1.8 V curve, a weak reduction current peak appeared, indicating a parasitic reaction between the electrolyte and electrode material. As the energy density increases with the working upper limit of the voltage range. As shown in Figure 6a, 6b, and 6c, the revealed biomass-derived carbon-based aqueous SSCs have a high specific capacitance (324 F g⁻¹ at 1 A g⁻¹) and excellent long cycling performance (90.6% retention after 50,000 cycles). Furthermore, a symmetric supercapacitor based on SPC-2 demonstrated the highest energy density (12.9 Wh kg⁻¹) and the highest power density of 8000 W kg⁻¹, showing promising application prospects.

3. CONCLUSION

We have employed facile pyrolysis and then an activation process to synthesize sulfur-doped porous carbon. The SPC materials with a high specific surface area (up to 2490 m² g⁻¹) have a high sulfur doping content. The optimized SPC-2 material displays the largest specific capacity (324 F g⁻¹ at 1 A g⁻¹) and excellent long cycling performance (90.6% retention after 50,000 cycles). Furthermore, a symmetric supercapacitor based on SPC-2 demonstrated the highest energy density (12.9 Wh kg⁻¹) and the highest power density of 8000 W kg⁻¹, showing promising application prospects.

4. EXPERIMENTAL SECTION

4.1. Synthesis of the Mesoporous Carbon. Ullothrix was collected from the Anhui University of Science and Technology campus. First, the clean, dry ullothrix was pyrolyzed in an N₂ atmosphere at 500 °C for 1 h. After that, the samples were mixed and ground with KOH powder in a mortar with mass ratios of 1:1, 1:2, and 1:3. Then the mixtures were activated in the N₂ atmosphere at 800 °C for 1 h at a 3 °C min⁻¹ heating rate. After cooling to room temperature, the samples were washed several times with excess 2 M HCl solution and deionized water. Finally, the products were dried overnight in a vacuum at 120 °C. The as-prepared activated porous carbons were named SPC-1, SPC-2, and SPC-3.

4.2. Characterization of Materials. The SPC samples were characterized by using a powder X-ray diffractometer (Smartlab SE, Cu Kα radiation), SEM (Hitachi, Flex-SEM1000), and Raman spectroscopy (Renishaw, inVia). The specific surface areas of the samples were characterized by using a surface area and porosimetry analyzer (Gold APP Instruments Co. Ltd., V-Sorb 2800P). The SPC samples were also studied by using an X-ray photoelectron spectrometer (ThermoFisher Scientific, ESCALAB 250Xi).

4.3. Electrochemical Measurements. The working electrode was prepared by milling the SPC sample with poly(vinylidene fluoride) (binder, 10 wt %) in N-methyl-2-pyrrolidinone to form a homogeneous slurry. Then, the slurry was spread on a piece of Ni foam (1 × 1 cm²) and dried under vacuum at 80 °C overnight. After that, a Hg/HgO electrode and a Pt foil were used as the reference electrode and counter electrode in a 2 M KOH electrolyte.
Cyclic voltammetry (CV), chronopotentiometry (CP), and electrochemical impedance spectroscopy (EIS) tests were conducted on a CH1660E electrochemical workstation to evaluate the electrochemical performance of the electrode. The cycling stability performance tests were conducted on a CT2001A LAND battery test system. The qualities of active materials were acquired from the mass change of the Ni foam, and the average loading mass was about 2 mg cm$^{-2}$.

A symmetric supercapacitor was fabricated by using the SPC as both positive and negative electrodes. A piece of cellulose paper was used as a separator, and a 2 M KOH solution was used as the electrolyte. The specific capacitance of the symmetric supercapacitor was calculated on the basis of the total mass of the active materials, and the calculation formulas of the specific capacitance, energy density, and power density are shown in the Supporting Information.

**ASSOCIATED CONTENT**

- Supporting Information
  The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.1c06253.
  Equations of specific capacitance, energy density, and power density; $N_2$ adsorption/desorption isotherms and pore size distributions of SPC; CV curves of SPC-1 and SPC-3; GCD curves of SPC-1 and SPC-3; and EIS curve of SSS (PDF)

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**Notes**
The authors declare no competing financial interest.

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