“One-Step” Carbonization Activation of Garlic Seeds for Honeycomb-like Hierarchical Porous Carbon and Its High Supercapacitor Properties

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ABSTRACT: In this paper, a simple “one-step” route is introduced to prepare a kind of novel honeycomb-like hierarchical porous carbon (h-HPC) by carbonizing and activating garlic seeds. Due to its special microstructure, h-HPC shows excellent electrochemical properties and high supercapacitor performances. The experimental results reveal the following: (1) There exists an optimal condition for synthesizing h-HPC, i.e., 700 °C carbonization temperature and 1:1 mass ratio of KOH and garlic seeds. (2) h-HPC has a three-dimensional interconnected porous structure and exhibits a specific surface area as high as 1417 m²/g with a narrow pore size distribution. (3) When h-HPC is employed as an electrode material in supercapacitors, its specific capacitance reaches a value up to 268 F/g at a current density of 0.5 A/g and excellent rate capability. (4) The h-HPC-based symmetric supercapacitor shows a high energy density of 31.7 Wh/kg at a power density of 500 W/kg and retains 99.2% of the initial capacitance after 10,000 charge/discharge cycles at 200 mV/s. When compared with similar works, these data are competitive, which demonstrates that the garlic-derived h-HPC is a kind of promising electrode material for the next-generation high-energy-density supercapacitors.

1. INTRODUCTION

As efficient and environmentally friendly energy storage devices, supercapacitors (SCs) have distinct advantages over conventional capacitors and low-power Li-ion batteries.1–3 In recent years, SCs have attracted enormous attention due to fast charging rates, high power densities, excellent operation safety, and long cycle life.4 Even for high current density discharge, SCs can be quickly charged and have a long service life.5 Hence, SCs are expected for potential applications in portable electronics, electric vehicles, and other high-power areas. As a key SCs’ component, an adopted electrode material largely determines its overall electrochemical performance.

In general, an ideal SCs’ electrode material requires a high specific surface area and appropriate porous structures.5 Among various materials, porous carbons are the most widely used due to advantages including low cost, developed pore structure, diversity, super electrical conductivity, stable physical and chemical properties, environmental friendliness, and high capacitance performance.7,8 Regarding the specific surface area and pore architecture of carbonaceous materials, it has been well known that the micropores less than 2 nm in diameter with ionic accessibility can provide a large capacitance, while the mesopores (2–50 nm) and macropores (>50 nm) can provide pathways for ion transport, reducing the diffusion resistance and improving the rate performance of SCs.5,10 That is to say that it is crucial to develop hierarchical porous carbon (HPC) with interconnected pores in different scales.11 Constructing the HPC structures, as a promising energy storage material, plays a key role in improving the performance of the carbon-based SCs.12,13

Biomass is one kind of energy source, which generally refers to plants or plant-, animal-, and marine organism-derived materials. Because biomass is earth-abundant, renewable, and low cost, the preparation of porous carbons with biomass as a precursor is of many advantages in reducing cost, reducing environmental pollution, and achieving the maximum utilization of resources.14–16 In addition, as precursors to fabricate porous carbons, the natural raw materials with hierarchical structures, such as garlic seeds and bamboo, can offer wider ranges of pore scales and better topological structures than those using conventional porous materials with homogeneous pore sizes. In recent years, the high value reutilization of biomass-derived carbon has...
received extensive attention for applying in SC field due to its merits of renewability, unique microstructure, specific chemical composition and environment-friendly nature. Many researchers have studied the conversion of different biomass into porous carbons, such as broad bean shells, soybean root, banana, bamboo char, and rapeseed dregs. Wang et al. prepared a grape-based honeycomb-like porous carbon, and it exhibited a capacitance of 275 F/g at 0.5 A/g in a three-electrode cell. Liang et al. prepared a pomelo peel-derived 3D honeycomb-like porous carbon, which not only had a high specific capacitance of 342 F/g at 0.2 A/g but also showed a good cycling performance with 98% retention over 1000 cycles.

As a kind of cultivated plant, garlic is used both as food and for medicinal applications, which has rapid growth rate, short maturation cycle, and high yield. Garlic contains rich mineral elements, involving phosphorus (P), magnesium (Mg), calcium (Ca), iron (Fe), zinc (Zn), copper (Cu), manganese (Mn), selenium (Se), etc., which are very important for forming the biomass porous carbon and making the material more widely used. In addition, natural garlic can effectively absorb and transport ions and water molecules for its metabolism due to its well-connected microtexture and multichannel structures. Thus, garlic can be carbonized to synthesize biomass carbon with special interconnected, multichannel, and porous structures, which facilitate ion transport as well as electrolyte penetration and enhance energy storage capability.

In this paper, we choose garlic seeds as a precursor and propose a possible process to have a 3D porous carbon by using a “one-step” carbonization activation route. Experimental results reveal that the 3D porous carbon exhibits kinds of novel 3D interconnected pores and bimodal or multimodal pore size distributions (PSDs). The effects of the mass ratio of alkali (KOH) and garlic seeds and the carbonization temperature on the specific surface area and pore volume are carefully investigated. Moreover, electrochemical properties of h-HPC under the optimal experimental parameters are measured in detail. Obviously, compared to other research studies, the present work has several advantages: (1) Garlic seeds are resource-rich, low cost, and sustainable. (2) The process is environment-friendly without any toxic substance addition. (3) The process is simple and easy to handle, which makes it possible for industrial mass production. (4) The obtained h-HPC is of a large specific surface area, unique pore size distribution, and high electrical conductivity, which result in greatly enhanced electrochemical properties and show promising candidates for energy storage devices.

2. RESULTS AND DISCUSSION

Figure 1 shows the preparation process of h-HPC. It was noted that the carbonization temperature and KOH activation played important roles in development of the pore structures. Therefore, in order to optimize the reaction conditions, different mass ratios of KOH and garlic seeds, namely, 1:2 (HPC-0.5), 1:1 (HPC-1), 2:1 (HPC-2), and 3:1 (HPC-3), and various holding temperatures, namely, 600 °C (HPC-600), 700 °C (HPC-700), and 800 °C (HPC-800), were utilized in the experiment. For comparison, carbonized garlic seeds without KOH activation (GC) were also prepared in the same way.

2.1. Morphologies and Microstructures. Figure 2 shows the morphologies and microstructures of the ordinary carbonized garlic seeds (GC) and the “one-step” carbonization and activation samples (h-HPC). Obviously, they exhibited relatively dense and sponge-like porous microstructures. The KOH activation (mass ratio of KOH and garlic seeds was 1:1) resulted in not only a unique 3D highly interconnected hierarchical structure but also uniform distribution of microscopic pores. The reason was that during carbonization, KOH acted as a template and activation agent for forming the porous honeycomb-like structure. Moreover, a great number of pores originated from phase separation between pristine freeze-dried garlic seeds and water during pyrolysis, which was beneficial to the rapid transportation of electrons and the enhancement of electrochemical properties. Furthermore, TEM observation reveals a great number of nanopores with circle-like outlines in the h-HPC samples, while it is not in the GC sample. That was to say that the honeycomb-like porous structure was formed during “one-step” carbonization and activation.
As we all know, carbonization temperature and the mass of KOH during activation are key factors in forming the pore structure. Figures S1 and S2 show the SEM morphologies of h-HPC at different carbonization temperatures and mass ratios of KOH and garlic seeds. It was found that the specific surface area reached the largest value at 700 °C and an optimum mass ratio of 1:1, while the excessive temperature and the other mass ratios led to the immoderate pore structures.

Figure 3a illustrates the Raman spectra of GC and h-HPC. Obviously, D and G bands were located at 1350 and 1620 cm⁻¹, respectively, where the D band represented the disorder from structural defects and impurities and the G band was generated from internal vibrations of the sp²-hybridized carbon.11 In general, the intensity ratio of D band to G band (I_D/I_G) gives the degree of crystallinity of carbon materials. Herein, the I_D/I_G values of h-HPC and GC were 1.03 and 0.87, respectively, indicating that KOH activation induced more defects in carbon sheets, further confirming the developed porous structure of h-HPC. The XRD patterns, as shown in Figure 3b, exhibited two peaks at 25° and 43.5° corresponding to the characteristic (002) and (101) crystalline planes. The low peak intensity and wide peaks revealed the amorphous structure of the samples.20,22

Figure 4. XPS spectra of h-HPC: (a) wide-range and (b) high-resolution C 1s.

2.2. Nitrogen Adsorption/Desorption Isotherm and Pore Size Distribution. Figure 5 illustrates the nitrogen adsorption/desorption isotherms (NADI) and the pore width distributions (PSDs) of GC and h-HPC. From Table 1, in the

Table 1. Geometric Parameters of the h-HPC and GC

| sample | S_BET (m²/g) | V_total (cm³/g) | d (nm) |
|--------|--------------|----------------|--------|
| h-HPC  | 1417.1       | 0.87           | 2.44   |
| GC     | 1.1          | 0.006          | 9.12   |

^a^Total surface: S_BET; total pore volume: V_total; average pore diameter: d_c.
case of the optimal h-HPC sample (HPC-700) with a mass ratio of 1:1 of KOH and garlic seeds, the NADI exhibited the largest specific surface area \( (S_{BET}) \) of 1417.1 m\(^2\)/g and the pore width distribution was of a total pore volume of 0.87 cm\(^3\)/g. However, for the GC sample, the \( S_{BET} \) was only 1.1 m\(^2\)/g and the total pore volume was 0.01 cm\(^3\)/g. In addition, a large number of pores were obtained on the cross section of the h-HPC (HPC-700) sample, as shown in Figure 5b, which contributed to the reaction between the activator and the internal region.27

2.3. Electrochemical Properties. In order to evaluate the electrochemical property of the h-HPC electrode, a two-electrode symmetric SC was prepared, and the test was performed in 6 mol/L KOH electrolyte. As shown in Figure 6, the experimental results revealed the following:

1. The specific capacitance reaches up to 228.2 F/g at 0.5 A/g when the scan rates vary from 5 to 200 mV/s and the voltage window from 0 to 1 V.

2. The symmetric SC CV curves also maintained a relatively rectangular shape without any deformation at other scan rates, which indicates that the double-layer capacitance was beneficial to the high specific capacitance.

3. The Ragone plot of the h-HPC symmetric SCs demonstrated that energy density was up to 31.7 Wh/kg and power density was 500 W/kg.

4. After 10,000 cycles, the h-HPC symmetric SCs retain 99.2% of the initial capacitance at a scan rate of 200 mV/s, which meant excellent cycling stability. The degradation of the cyclic performance during charging and discharging was ascribed to the reasons including porous structural collapse, coupled with the immersion of electrolyte, changes in the microstructures and specific surface area, etc.

5. Electrochemical impedance spectroscopy (EIS) measurement indicated that the Nyquist plot of the electrode exhibited almost a vertical straight line in a low frequency range, which implied the low-grade ion diffusion resistance of the 3D h-HPC. Furthermore, Figure S4 illustrates the Nyquist plots of the h-HPC and GC electrodes in three-electrode symmetric SCs in 6 mol/L KOH electrolyte. Obviously, h-HPC provided a fast ion charge transfer, resulting in excellent SC performance.

In addition, the three-electrode electrochemical properties of h-HPC were measured in 6 mol/L KOH solution, as shown in Figure 7. Owing to the 3D porous structure and extremely high specific surface area, the h-HPC electrode was of also high SC properties. The main results were as follows:

1. The CV curves showed a typical rectangular shape, which could be maintained even at 200 mV s\(^{-1}\), confirming the contribution of electrical double-layer capacitance.

2. The galvanostatic charge/discharge curves exhibited a desirable linearity and triangular symmetry, which
indicated the superior electrochemical reversibility and coulombic efficiency.

(3) The significant voltage drop did not appear even at 10 A/g due to low internal resistance.

(4) The specific capacitance calculated from the charge/discharge curves was up to 268 F/g at 0.5 Ag, which was almost five times higher than that of the GC electrode (56 F/g at 0.5 A/g).

(5) More importantly, even at an extremely high current density of 20 A/g, the h-HPC electrode still had a high capacitance of 214 F/g.

The superior charge storage behavior of h-HPC could be ascribed to the following reasons:

(1) The small pore size (<1 nm) led to a super specific surface area and provided abundant electroactive sites for SCs.

(2) On the one hand, a great number of micropores supplied a cumulative space for electrons; on the other hand, the mesopores shortened the distance of ion diffusion, which was beneficial to buffer ion penetration into the inner micropores; and last, the macropores acted as the storage layer. Also, the continuous pathway or ion-buffering container was formed because of the interconnected honeycomb-like porous structure, which resulted in a rapid ionic transport.

(3) The super electrical conductivity of h-HPC and good wettability of the oxygen functional groups also contributed to the specific capacity and rate performance.

2.4. Effect of the Carbonization Temperature. Table S1, Table S2, and Figure S5 give the NADI and pore width distribution curves of the h-HPC samples at different carbonization temperatures and various mass ratios of KOH and garlic seeds. It was found that the optimal value was 700 °C, and the specific surface area aggravated with an increase in reaction temperature initially. This phenomenon could be ascribed to the consumption of carbon atoms and the formation of pores due to the availability of sufficient energy.

In fact, during carbonization, especially the KOH activation, the process included chemical reactions, such as conversion of K compounds into metallic K, oxidation of C to CO/CO2 or carbonate, and a series of reactions among various activations. That followed the following steps, i.e., (1) the chemical reactions started with eq(1) and formed K2CO3 at 400−600 °C; (2) K2CO3 decomposed into K2O and CO2 when the reaction temperature was higher than 700 °C (eq (2)); (3) the generated CO2 and K compounds further reacted with garlic seed-derived carbon when the temperature exceeded 700 °C (eqs (3)−(5)); and (4) in the end, KOH entered these pores and reacted with the garlic

Figure 7. Electrochemical properties of the h-HPC electrode in a three-electrode system in 6 mol/L KOH electrolyte: (a) CV curves at different scan rates (5−200 mV/s); (b) galvanostatic charge/discharge curves at different current densities (0.5−20.0 A/g); (c) CV curves at a scan rate of 50 mV/s; (d) galvanostatic charge−discharge curves at a current density of 1.0 A/g; (e) specific capacitance at different current densities; and (f) cyclic performance at a scan rate of 200 mV/s (inset: the 1st and 10,000th cycle CV curves).
seed-derived carbonized substance, resulting in the high specific surface area and honeycomb-like hierarchical porous carbon (hHPC).\(^1,^{10}\)

\[
\begin{align*}
6\text{KOH} + 2\text{C} & \rightarrow 2\text{K} + 3\text{H}_2 + 2\text{K}_2\text{CO}_3 \\
\text{K}_2\text{CO}_3 & \rightarrow \text{K}_2\text{O} + \text{CO}_2 \\
\text{CO}_2 + \text{C} & \rightarrow 2\text{CO} \\
\text{K}_2\text{CO}_3 + 2\text{C} & \rightarrow 2\text{K} + 3\text{CO} \\
\text{K}_2\text{O} + \text{C} & \rightarrow 2\text{K} + \text{CO}
\end{align*}
\] (1-5)

Initially, with increasing temperature, more energy was provided for the reaction, and more carbon atoms at the active sites were consumed to form pores, which made the specific surface area and the pore volume gradually increase. However, when the temperature increased further, the excessive carbonization temperature would form the immoderate pores. In other words, the inappropriate activation reaction conditions destroyed the mechanical strength of the carbon skeleton. Especially, when the carbonization temperature exceeded 700 °C, the pore structures radically collapsed and the specific surface area and pore volume cut down sharply.

Figure 8 illustrates the electrochemical properties of the hHPC electrode at different carbonization temperatures in 6 mol/L KOH electrolyte. The specific capacitances of the hHPC electrode, which was carbonized at 600, 700, and 800 °C, were 248.5, 268, and 189 F/g (0.5 A/g), respectively. Obviously, these data showed the similar variation of the specific surface areas at different carbonization temperatures.

2.5. Effect of the Mass Ratio of KOH and Garlic Seeds.

Figure 9 illustrates the electrochemical properties of the hHPC electrode at different mass ratios of KOH and garlic seeds in 6 mol/L KOH electrolyte. The specific capacitances of hHPC at mass ratios of 1:2 (HPC-0.5), 1:1 (HPC-1), 2:1 (HPC-2), 3:1 (HPC-3), and 4:1 (HPC-4) were calculated as 218.5, 268, 257.7, 249.5, and 176.4 F/g (0.5 A/g), respectively. In regard to the morphology changes, as shown in Figure S2, the desired porous structure and large specific surface area could only be formed in higher KOH contents. However, the experiment revealed that the optimal condition was the mass ratio 1:1, which not only had the largest specific surface area of 1417 m\(^2\)/g but also exhibited the highest specific capacitance of 268 F/g.

Totally, Figure 10 gives the influence of process conditions on specific capacitance of the hHPC electrode in 6 mol/L KOH electrolyte. Table 2 lists the data comparisons of capacitance and energy density with other research works by using various carbonaceous materials.\(^5,^{6,17,23,30-35}\) Obviously, the present work showed excellent properties.

3. CONCLUSIONS

A kind of 3D honeycomb-like hierarchical porous carbon (hHPC) is successfully synthesized by using garlic seeds as a precursor via a simple “one-step” route, i.e., freeze-drying garlic seeds into powder, then carbonization, and activation. Due to its special microstructure, hHPC exhibits a high specific surface area up to 1417 m\(^2\)/g and a narrow pore size distribution under the optimal conditions, i.e., 700 °C carbonization temperature and 1:1 mass ratio of KOH and garlic seeds. It is also demonstrated that the hHPC electrode shows excellent electrochemical properties and high supercapacitor performances, such as a specific capacitance of 268 F/g at a current density of 0.5 A/g, energy density of 31.7 Wh/kg, and power density of 500 W/kg, and retains 99.2% of the initial capacitance after 10,000 charge/discharge cycles at 200 mV/s. These data are higher than those of the most other reported carbon-based electrodes. This process is of advantages including simple, economic, environment-friendly, applicable for mass production, etc. and is expected to be broadly applied as a promising electrode material for the next-generation high-energy-density supercapacitors.

4. EXPERIMENTAL SECTION

Garlic seeds were purchased from Qi County, Henan Province, China. HCl (36.0–38.0%), and KOH were from Aladdin Industrial Corporation. Nickel foam, polytetrafluoroethylene (PTFE), and carbon black were from a reputed source. Deionized water was used during the experimental process.

Figure 1 schematically illustrates the synthesis process of hHPC. The detailed steps were as follows:

1. A total of 100 g of fresh garlic seeds was washed with distilled water several times in order to remove the impurities.
The garlic seeds were put in a freeze-drier and kept for 56 h to create a porous structure due to ice sublimation. The dried garlic seeds (about 2.75 g) were ground into powder and then mixed with equivalent mass of KOH in 75 mL of distilled water. The above mixture solution was stirred vigorously for 1 h and dried at 80 °C for 48 h. The above product was heated to 700 °C with a heating rate of 5 °C/min and held for 2 h in a tube furnace with a constant flow of pure argon to get the intermediate product of h-HPC. The intermediate product was washed with a 0.1 mol/L HCl solution, distilled water, and ethanol repeatedly in order to remove the remaining potassium species. Last, the above “one-step” carbonization and activation samples were dried in a vacuum-drying chamber at 80 °C for 12 h, obtaining h-HPC.

In the experiment, different mass ratios of KOH and garlic seeds, namely, 1:2 (HPC-0.5), 1:1 (HPC-1), 2:1 (HPC-2), and 3:1 (HPC-3), and various holding temperatures, namely, 600 °C (HPC-600), 700 °C (HPC-700), and 800 °C (HPC-800), were utilized in order to optimize the reaction conditions. For comparison, the carbonized garlic seeds without KOH activation (GC) were also prepared in the same way.

The morphologies and microstructures of the samples were characterized by using analytical instruments, including a scanning electron microscope (SEM, S-4800, Hitachi, Japan), transmission electron microscope (TEM, JEM-2100, JEOL Ltd., Japan), X-ray diffractometer (XRD, D8 Advance, Bruker AXS, Germany) equipped with Cu Kα radiation, and scanning confocal micro-Raman spectrometer (LabRAM HR, HORIBA, France) with a laser excitation wavelength of 488 nm and recorded in a range of 1000−3000 cm⁻¹.

The surface chemical compositions were measured by X-ray photoelectron spectroscopy (XPS, ESCALAB 250Xi, Thermo Fisher, USA). The specific surface area and pore volumes were measured in nitrogen adsorption/desorption (JW-BK 122 W, JWGB, China) at 77 K.

The electrochemical properties of the samples were determined regarding the following steps, principles, and processes. These were as follows:

1. A mixed slurry of h-HPC, carbon black, and poly(tetrafluoroethylene) was prepared with a 8:1:1 mass ratio in ethanol.
2. After drying at 80 °C for 24 h, the sample was subsequently coated with nickel (Ni) foam (1 cm × 1 cm) and punched in a tablet press under 13 MPa for making the SC electrode.
3. Each electrode was loaded with h-HPC, which was measured to be 4.0 mg/cm².
4. The electrochemical tests of the SC electrodes were adopted in 6.0 mol/L KOH electrolyte. For a three-

![Figure 9](image_url)  
**Figure 9.** Electrochemical properties of the h-HPC electrode in 6 mol/L KOH electrolyte at different mass ratios of HPC-0.5 (1:2), HPC-1 (1:1), HPC-2 (2:1), and HPC-3 (3:1) of KOH and garlic seeds: (a) CV curves at a scan rate of 50 mV/s; (b) galvanostatic charge−discharge curves at a current density of 1.0A/g. (c) Experimental Nyquist plot over the frequency range from 0.01 Hz to 100 kHz.

![Figure 10](image_url)  
**Figure 10.** Influence of process conditions on specific capacitance of the h-HPC electrode in 6 mol/L KOH electrolyte: (a) carbonization temperatures and (b) mass ratios.
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discharge measurements by using the following equation:

\[ C = \frac{I \times \Delta t}{m \times \Delta V} \]  

where \( I \) is the discharge current (A), \( m \) is the mass of active materials in an electrode (g), \( \Delta t \) is the discharge time (s), and \( \Delta V \) is the potential window vs (Hg/HgO) (V).

The gravimetric specific capacitance \((C, \text{F/g})\) values of the supercapacitor electrode also were calculated from cyclic voltammetry (CV) measurements by using the following equation:

\[ C = \frac{\int I \times dV}{\nu \times m \times V} \]  

where \( I \) is the current in CV curves (A), \( V \) is the potential scan rate (V/s), \( \nu \) is the potential scan rate (V/s), and \( m \) is the mass of the active materials in an electrode (g).

For the two-electrode system, the gravimetric specific capacitance was calculated by the equation

\[ C = 2(I \times \Delta t)/(m \times \Delta V) \]  

where \( I \) is the discharge current (A), \( \Delta t \) is the discharge time (s), \( \Delta V \) is the potential window (V), and \( m \) is the mass of the active materials in a single working electrode (g).

The energy density \((E, \text{Wh/kg})\) and power density \((P, \text{W/kg})\) of supercapacitors were calculated by the following equations:

\[ E = \frac{(C \times V^2)}{2} \]  

\[ P = \frac{E}{t} \]  

where \( V \) is the voltage window of the supercapacitor (V) and \( t \) is the discharge time (s).

### Table 2. Comparisons of the Capacitance and Energy Density with Other Reports

| materials                     | \( C \) (F/g) | current density (A/g) | electrolyte | \( E \) (Wh/kg) | \( S_{\text{BET}} \) (m\(^2\)/g) | cyclic performance | ref.   |
|-------------------------------|--------------|-----------------------|-------------|---------------|-------------------------------|----------------------|-------|
| graphene/carbon black         | 135          | 10                    | 6 mol/L KOH | 5             | 96.1% (2000 cycles)           | 30                   |
| macroporous graphene          | 202          | 1                     | 1 mol/L Na\(_2\)SO\(_4\) | 5             | 95% (2000 cycles)            | 31                   |
| N-doped carbon fiber          | 202          | 1                     | 6 mol/L KOH | 7.1           | 562.51 (3000 cycles)         | 32                   |
| hexaporous carbon             | 154          | 2                     | 1 mol/L H\(_2\)SO\(_4\) | 5.3           | 1720 (1000 cycles)           | 33                   |
| grape-based porous carbon     | 275          | 0.5                   | 6 mol/L KOH | 12.6          | 1268 (5000 cycles)           | 8                    |
| seaweed-derived carbon        | 198          | 1                     | 1 mol/L H\(_2\)SO\(_4\) | 7.4           | 3500 (85% (1000 cycles)      | 34                   |
| pomelo peel                   | 342          | 10                    | 6 mol/L KOH | 9.4           | 2725 (98% (1000 cycles)      | 23                   |
| bamboo char                   | 222          | 0.5                   | KOH/PVA     | 20.6          | 1732 (84% (5000 cycles)      | 17                   |
| banana                        | 178.9        | 5                     | 6 mol/L KOH | 3.32          | 1414.97 (98% (10,000 cycles) | 6                    |
| almond-derived                | 228          | 1                     | 6 mol/L KOH | 9.4           | 2725 (98% (1000 cycles)      | 35                   |
| h-HPC from garlic seeds       | 268          | 0.5                   | 6 mol/L KOH | 31.7          | 1417.1 (99.2% (10,000 cycles) | this work            |

(5) The two-electrode symmetric SC was built with h-HPC, while the mass loading of each SC electrode was 4.0 mg/cm\(^2\), and the test was then performed in 6.0 mol/L KOH solution. Before testing, the SC electrodes were vacuum-impregnated in a two-electrode cell in electrolyte for 5 h.

All the above experiments were carried out on an electrochemical workstation. The electrochemical impedance spectroscopy (EIS) was implemented at an AC amplitude of 5 mV with the frequency from 0.1 Hz to 100 kHz.

The gravimetric specific capacitance \((C, \text{F/g})\) values of the electrode were calculated based on galvanostatic charge/dischage measurements by using the following equation:

\[ C = \frac{I \times \Delta t}{(m \times \Delta V)} \]  

where \( I \) is the discharge current (A), \( m \) is the mass of active materials in an electrode (g), \( \Delta t \) is the discharge time (s), and \( \Delta V \) is the voltage window of the supercapacitor electrode also were calculated from cyclic voltammetry (CV) measurements by using the following equation:

\[ C = \frac{\int I \times dV}{(\nu \times m \times V)} \]  

where \( I \) is the current in CV curves (A), \( V \) is the potential scan rate (V/s), \( \nu \) is the potential scan rate (V/s), and \( m \) is the mass of the active materials in an electrode (g).

(5) The two-electrode symmetric SC was built with h-HPC, while the mass loading of each SC electrode was 4.0 mg/cm\(^2\), and the test was then performed in 6.0 mol/L KOH solution. Before testing, the SC electrodes were vacuum-impregnated in a two-electrode cell in electrolyte for 5 h.

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### SUPPORTING INFORMATION

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.0c04190.

SEM morphologies, geometric parameters, NADI and pore width distribution curves of HPC at different carbonization temperatures and mass ratios of KOH and garlic seeds, high-resolution O 1s of h-HPC, Nyquist plots of the h-HPC and GC electrodes in a three-electrode symmetric SC in 6 mol/L KOH electrolyte, and coulombic efficacy (PDF)

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Notes
The authors declare no competing financial interest.

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