Electronic Supporting Information for

# Synthesis of Garlic Skin-Derived 3D Hierarchical Porous Carbon for High-Performance Supercapacitors

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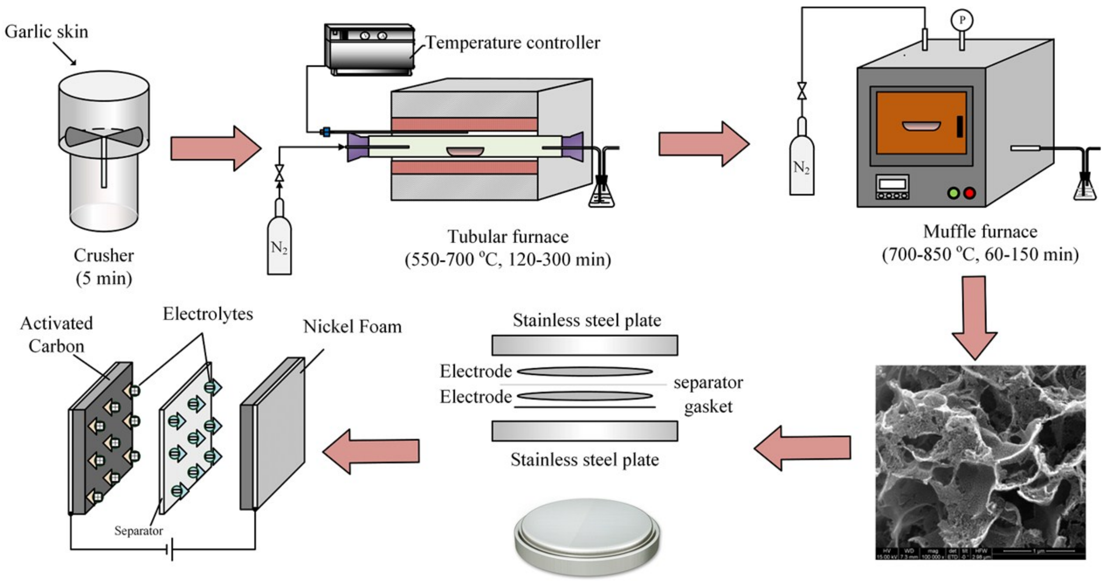


**Figure S1** Schematic illustration for the preparation of garlic-skin-derived hierarchical porous carbon.

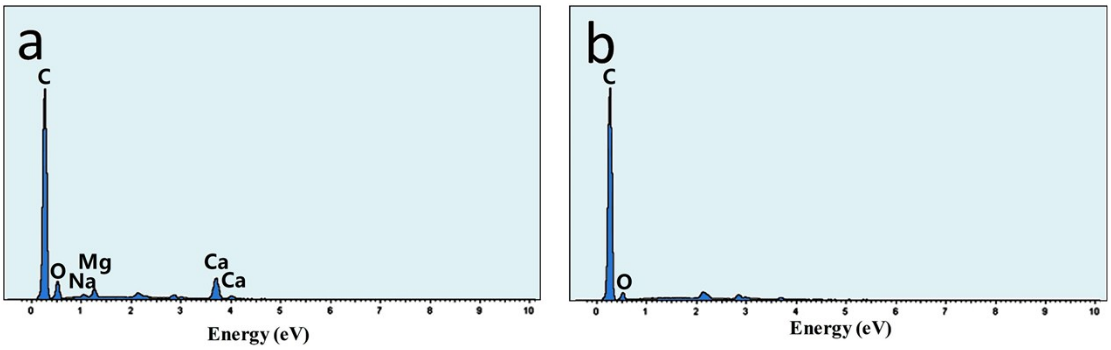


**Figure S2** (a) SEM-EDS image of GHCs before acid washing. (b) SEM-EDS image of GHCs after acid washing.

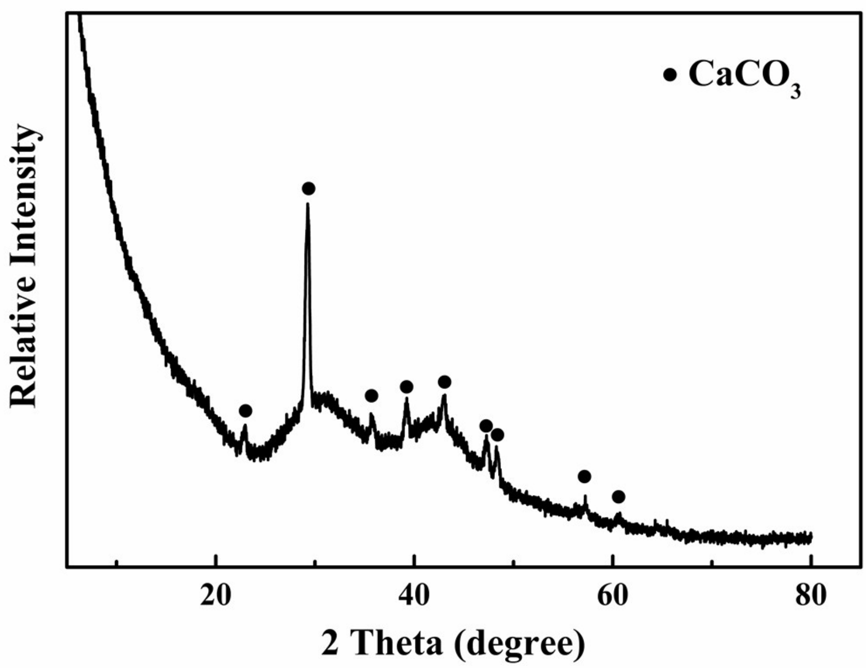


**Figure S3** XRD pattern of GHCs before acid washing.

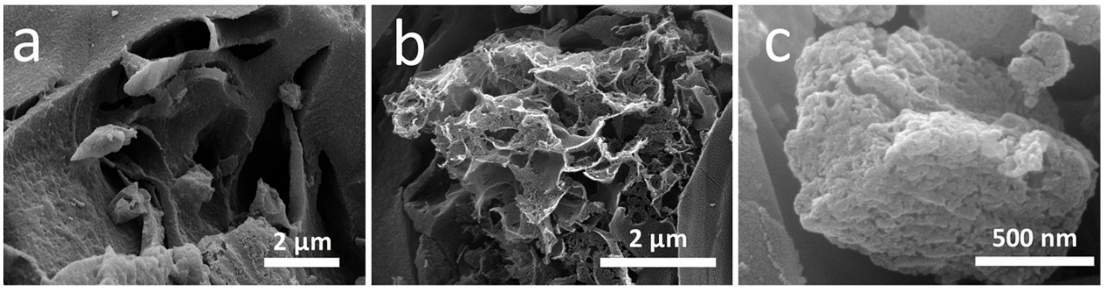


**Figure S4** SEM images of GHCs (a and b) before and (c) after acid washing.

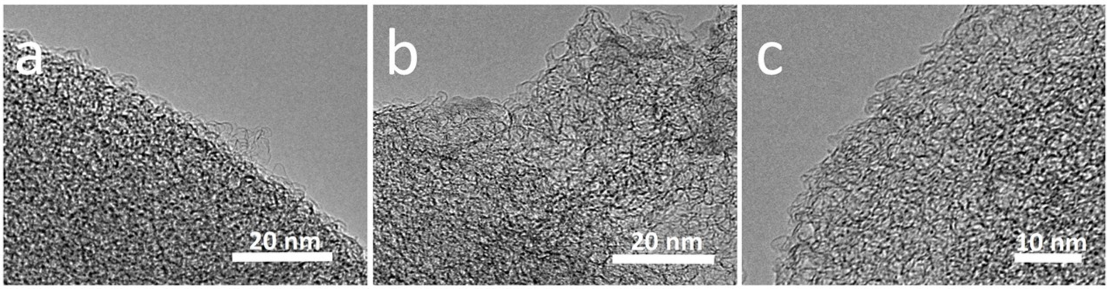


**Figure S5** TEM images of GHCs.

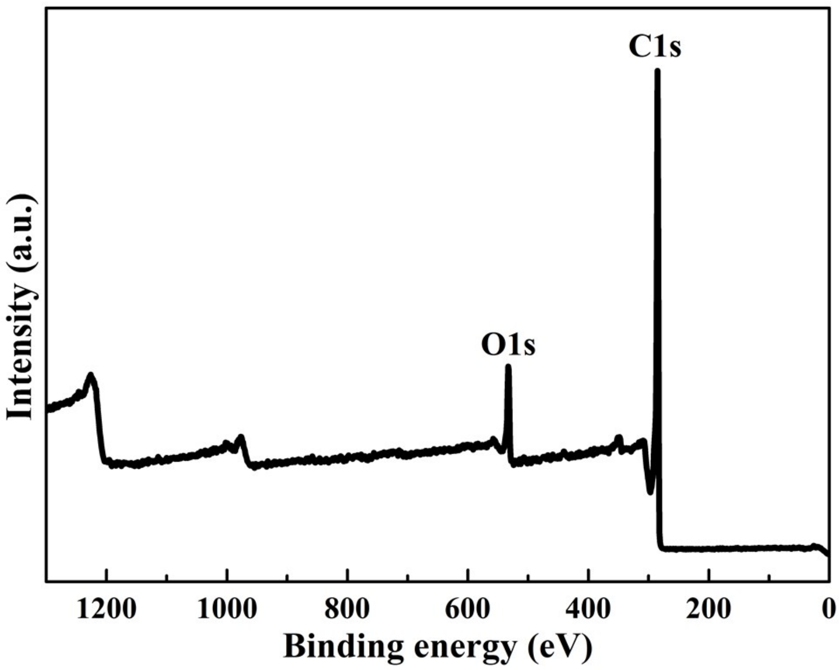


**Figure S6** X-ray photoelectron spectroscopy (XPS) analysis of GHC-17.

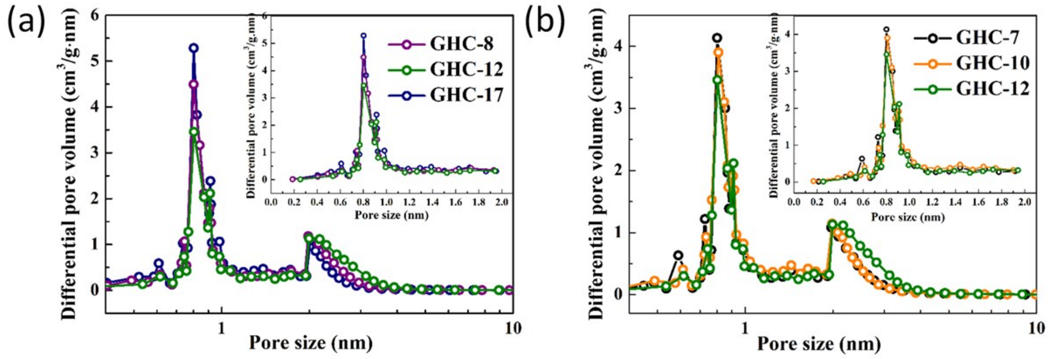


**Figure S7** Pore size distribution curve of (a) GHC-8, GHC-12, and GHC-17; (b) GHC-7, GHC-10, and GHC-12.

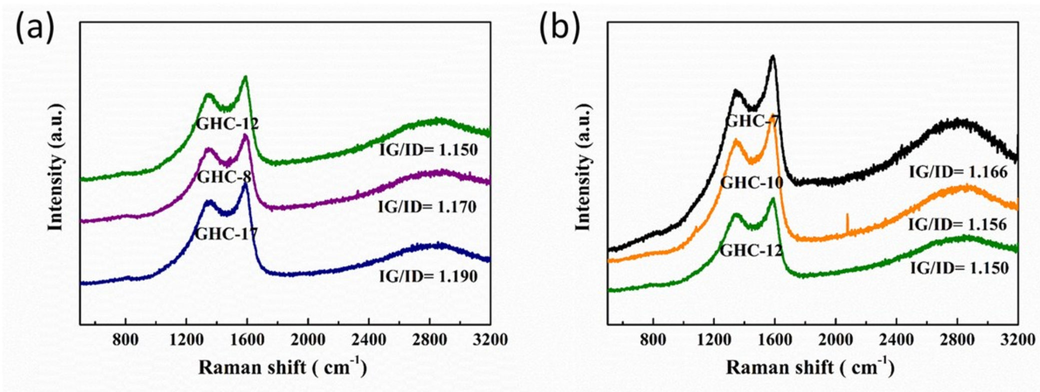


**Figure S8** (a) Raman spectrum of (a) GHC-8, GHC-12, and GHC-17; (b) GHC-7, GHC-10, and GHC-12.

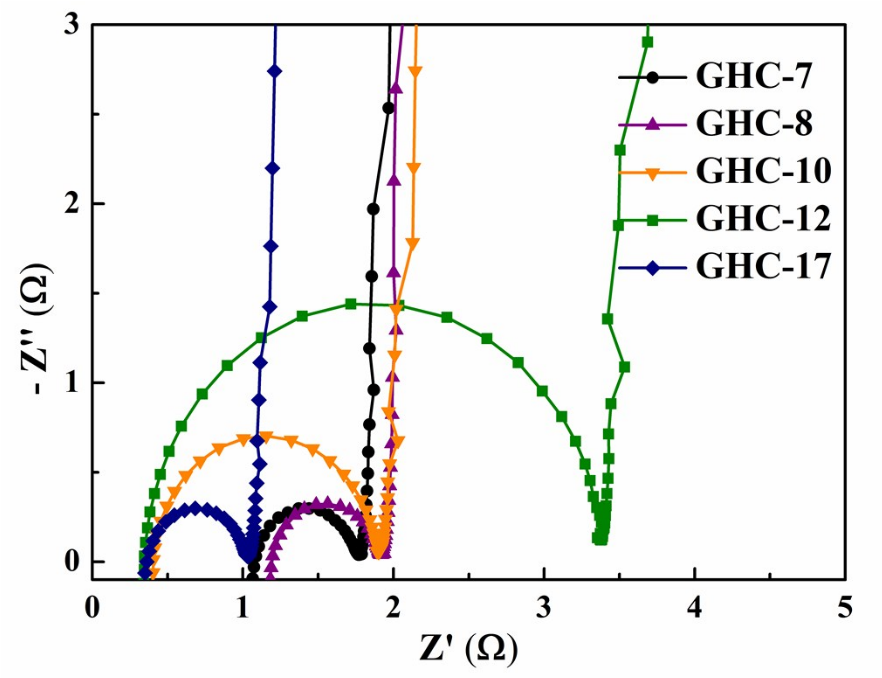


**Figure S9** Nyquist plots of GHC-7, GHC-8, GHC-10, GHC-12, and GHC-17.

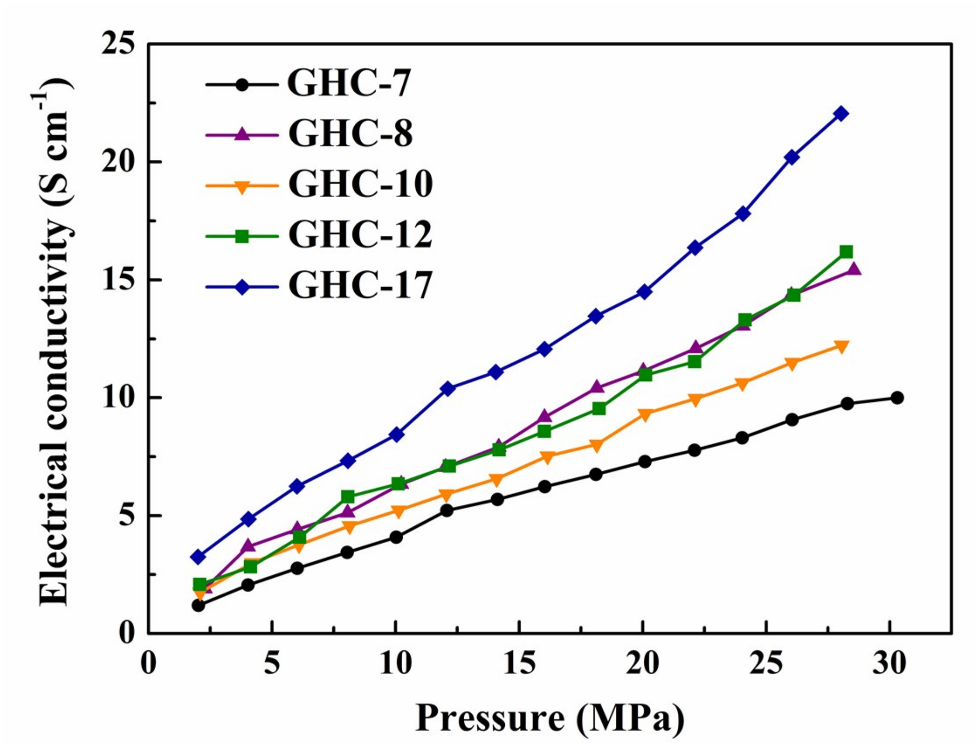


**Figure S10** The electrical conductivity of the GHC based electrodes measured by four-point method (25 $^{o}$C)

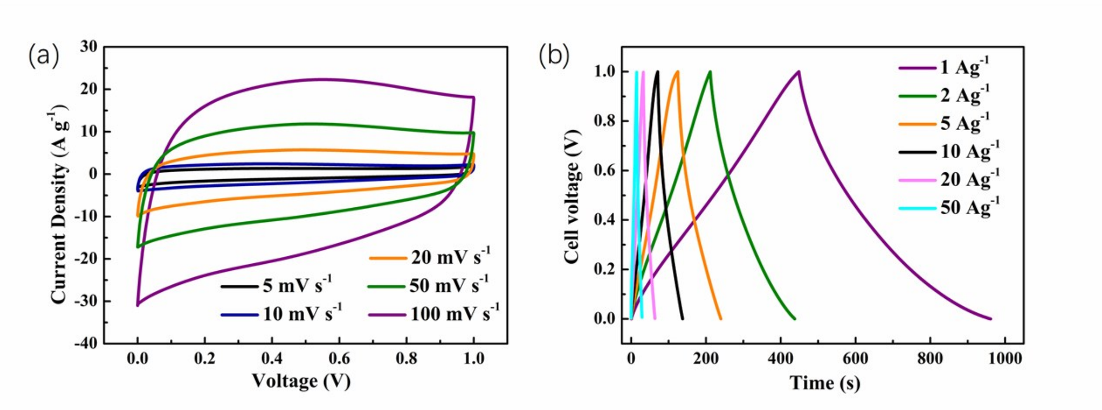


**Figure S11** Electrochemical performance of the GHC-17 sample measured in a three-electrode system using 6 mol·$L^{-1}$ KOH electrolyte. (a) CV curves of the GHC-17 at different scan rates from 5 to 100 mV·$s^{-1}$. (b) Charge-discharge curves of the GHC-17 at different current densities from 1 to 50 A·$g^{-1}$

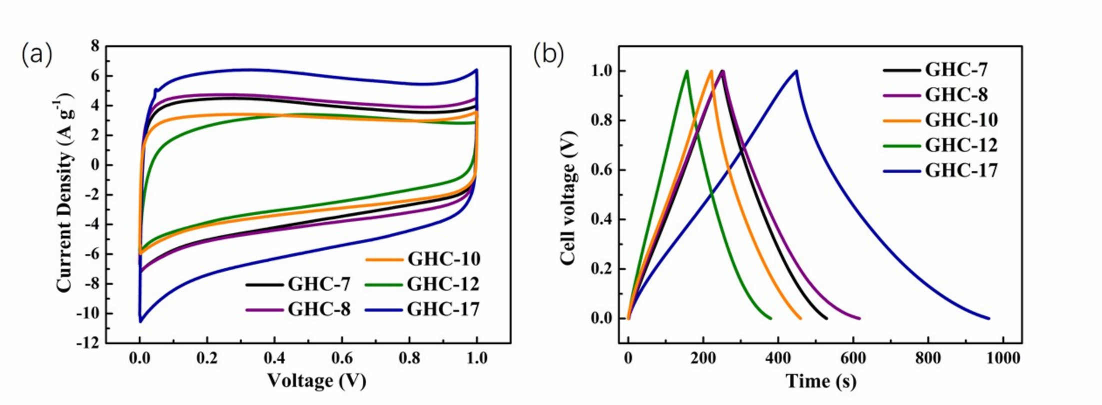


**Figure S12** Electrochemical performance of the GHC-7, GHC-8, GHC-10, GHC-12, GHC-17 samples measured in a three-electrode system using 6 mol $L^{-1}$ KOH as the electrolyte. (a) CV curves at 20 mV $s^{-1}$; (b) The galvanostatic charge-discharge profiles at 1 A $g^{-1}$

**Table S1** Factors and levels of the orthogonal design

| Levels | Factors | | | | |
|---|---|---|---|---|---|
| | Impregnation ratio A | Activation temperature B ($^oC$) | Activation time C (min) | Carbonization temperature D ($^oC$) | Carbonization time E (min) |
| **1** | 3.0:1 | 700 | 60 | 550 | 120 |
| **2** | 3.5:1 | 750 | 90 | 600 | 180 |
| **3** | 4.0:1 | 800 | 120 | 650 | 240 |
| **4** | 4.5:1 | 850 | 150 | 700 | 300 |

**Table S2** Orthogonal experimental arrangements

| Experiment No. | Range and levels | | | | |
|---|---|---|---|---|---|
| | Impregnation ratio A | Activation temperature B (°C) | Activation time C (min) | Carbonization temperature D ($^oC$) | Carbonization time E (min) |
| **1** | 3.0:1 | 700 | 60 | 550 | 120 |
| **2** | 3.0:1 | 750 | 90 | 600 | 180 |
| **3** | 3.0:1 | 800 | 120 | 650 | 240 |
| **4** | 3.0:1 | 850 | 150 | 700 | 300 |
| **5** | 3.5:1 | 700 | 90 | 650 | 300 |
| **6** | 3.5:1 | 750 | 60 | 700 | 240 |
| **7** | 3.5:1 | 800 | 150 | 550 | 180 |
| **8** | 3.5:1 | 850 | 120 | 600 | 120 |
| **9** | 4.0:1 | 700 | 150 | 700 | 180 |
| **10** | 4.0:1 | 750 | 60 | 650 | 120 |
| **11** | 4.0:1 | 800 | 90 | 600 | 300 |
| **12** | 4.0:1 | 850 | 90 | 550 | 240 |
| **13** | 4.5:1 | 700 | 150 | 600 | 240 |
| **14** | 4.5:1 | 750 | 120 | 550 | 300 |
| **15** | 4.5:1 | 800 | 90 | 700 | 120 |
| **16** | 4.5:1 | 850 | 60 | 600 | 180 |

**Table S3** Specific surface area and pore volume at different fabrication conditions

| Experiment No. | Variables | | | | | Responses | |
|---|---|---|---|---|---|---|---|
| | A | B | C | D | E | Specific surface area ($m^2/g$) | Pore volume ($cm^3/g$) |
| **1** | 1 | 1 | 1 | 1 | 1 | 1468.33 | 0.759 |
| **2** | 1 | 2 | 2 | 2 | 2 | 1701.82 | 0.896 |
| **3** | 1 | 3 | 3 | 3 | 3 | 1714.46 | 0.922 |
| **4** | 1 | 4 | 4 | 4 | 4 | 1568.98 | 0.852 |
| **5** | 2 | 1 | 2 | 3 | 4 | 1725.09 | 0.938 |
| **6** | 2 | 2 | 1 | 4 | 3 | 1442.16 | 0.795 |
| **7** | 2 | 3 | 4 | 1 | 2 | 2268.66 | 1.162 |
| **8** | 2 | 4 | 3 | 2 | 1 | 2542.77 | 1.378 |
| **9** | 3 | 1 | 3 | 4 | 2 | 1561.91 | 0.838 |
| **10** | 3 | 2 | 4 | 3 | 1 | 2259.80 | 1.218 |
| **11** | 3 | 3 | 1 | 2 | 4 | 2077.21 | 1.165 |
| **12** | 3 | 4 | 2 | 1 | 3 | 2254.06 | 1.336 |
| **13** | 4 | 1 | 4 | 2 | 3 | 1886.07 | 1.063 |
| **14** | 4 | 2 | 3 | 1 | 4 | 1507.58 | 0.865 |
| **15** | 4 | 3 | 2 | 4 | 1 | 1161.90 | 0.664 |
| **16** | 4 | 4 | 1 | 3 | 2 | 1470.07 | 0.824 |
| **17** | 3 | 3 | 4 | 2 | 1 | 2818.22 | 1.327 |

**Table S4** Range analysis results

| Indicators | Levels | Factors | | | | |
|---|---|---|---|---|---|---|
| | | **A** | **B** | **C** | **D** | **E** |
| **Specific surface area $S_{BET}$** | $K_{1j}$ | 1613.40 | 1660.35 | 1614.44 | 1874.66 | 1858.20 |
| | $K_{2j}$ | 1994.67 | 1727.84 | 1710.72 | 2051.97 | 1750.62 |
| | $K_{3j}$ | 2038.25 | 2194.99 | 1831.68 | 1792.36 | 1824.19 |
| | $K_{4j}$ | 1506.41 | 1958.97 | 1995.88 | 1433.74 | 1719.72 |
| | $R_j$ | 531.84 | 534.64 | 381.44 | 618.23 | 138.49 |
| | **The optimal solution** | $A_3$ | $B_3$ | $C_4$ | $D_2$ | $E_1$ |
| | **Levels** | **Factors** | | | | |
| | | A | B | C | D | E |
| **Pore volume** | $K_{1j}$ | 0.86 | 0.90 | 0.89 | 1.03 | 1.03 |
| | $K_{2j}$ | 1.07 | 0.94 | 0.96 | 1.13 | 0.93 |
| | $K_{3j}$ | 1.14 | 1.20 | 1.00 | 0.98 | 1.00 |
| | $K_{4j}$ | 0.85 | 1.10 | 1.07 | 0.79 | 0.96 |
| | $R_j$ | 0.29 | 0.30 | 0.19 | 0.34 | 0.10 |
| | **The optimal solution** | $A_3$ | $B_3$ | $C_4$ | $D_2$ | $E_1$ |

**Table S5** Characteristics of garlic skin-derived activated carbons without KOH activation
(Samples are prepared with carbonization time for 2h)

| Experiment No. | Carbonization temperature D ($^{o}C$) | Specific surface area ($m^2/g$) | Pore volume ($cm^3/g$) | Average pore diameter (nm) |
|---|---|---|---|---|
| **1-1** | 550 | 81.44 | 0.079 | 3.812 |
| **1-2** | 600 | 178.16 | 0.143 | 3.315 |
| **1-3** | 650 | 266.48 | 0.216 | 2.947 |
| **1-4** | 700 | 420.61 | 0.251 | 2.759 |

**Table S6** Peak assignment of C 1s and O 1s of the GHC-17

| Peak | Binding energy (eV) | Assignment | Fraction of species | Atom (%) |
|---|---|---|---|---|
| **C 1s** | 284.7 | $Sp^3C=C$ | 32.7 | 87.08 |
| | 285.6 | $Sp^2C-C$ | 23.2 | |
| | 286.5 | C-O | 21.4 | |
| | 288.3 | C-OOH | 22.7 | |
| **O 1s** | 531.5 | C=O Cabonyl O in COOR | 25.0 | 12.92 |
| | 532.4 | C-OH, C-O-C | 25.3 | |
| | 533.8 | Non-carbonyl O in COOR | 25.2 | |
| | 535.5 | $H_2O_{(ads)}/O_{2(ads)}$ | 24.6 | |

**Table S7** Pore parameters of GHC-7, GHC-8, GHC-10, GHC-12, GHC-17 ($S_{BET}$: total surface area; $S_{micro}$: micropore specific surface area; $S_{meso}$: mesoporous specific surface area; $V_T$: total pore volume; $V_{micro}$: micropore volume; $V_{meso}$: mesoporous volume; $d_a$: average pore diameter)

| Sample | $S_{BET}$ ($m^2$ $g^{-1}$) | $S_{micro}$ ($m^2$ $g^{-1}$) | $S_{meso}$ ($m^2$ $g^{-1}$) | $V_{tot}$ ($cm^3$ $g^{-1}$) | $V_{micro}$ ($cm^3$ $g^{-1}$) | $V_{meso}$ ($cm^3$ $g^{-1}$) | $d_a$ (nm) | Capacitance (F $g^{-1}$) |
|---|---|---|---|---|---|---|---|---|
| **7** | 2268 | 1606 | 662 | 1.162 | 0.687 | 0.475 | 2.049 | 245 |
| **10** | 2260 | 1311 | 949 | 1.218 | 0.535 | 0.683 | 2.008 | 207 |
| **12** | 2254 | 851 | 1403 | 1.336 | 0.366 | 0.970 | 2.355 | 173 |
| **8** | 2543 | 1721 | 822 | 1.378 | 0.808 | 0.570 | 2.055 | 268 |
| **17** | 2818 | 2422 | 396 | 1.327 | 1.022 | 0.305 | 1.883 | 427 |

**Table S8** The electrical conductivity of the GHC based electrodes measured by four-point method (25 $^{o}$C)

| | **Electrical conductivity (S $cm^{-1}$)** | |
|---|---|---|
| | **2 MPa** | **28 MPa** |
| **GHC-7** | 1.19 | 9.74 |
| **GHC-8** | 1.9 | 15.17 |
| **GHC-10** | 1.74 | 12.22 |
| **GHC-12** | 2.08 | 16.18 |
| **GHC-17** | 3.25 | 22.05 |

**Table S9** Summary of capacitive performance of carbon-based supercapacitor electrodes

| Carbon material | $S_{BET}$($m^2g^{-1}$) | Gravimetric capacitance (F $g^{-1}$) | Volumetric capacitance (F $cm^{-3}$) | Electrolyte & Test set-up | Ref. |
|---|---|---|---|---|---|

| | | | | | |
|---|---|---|---|---|---|
| **GHC** | 2818 | 427 (0.5 A $g^{-1}$) | 162 (0.5 A $g^{-1}$) | 6 M KOH Two electrode | This work |
| **Reed straw derived carbon** | 2387 | 355 (1 A $g^{-1}$) | -- | 6 M KOH Two electrode | 1 |
| **N-Doped carbon nanocages** | 1794 | 313 (1 A $g^{-1}$) | -- | 1 M KOH Two electrode | 2 |
| **Activated carbon fiber from sawdust** | 2395 | 242 (0.5 A $g^{-1}$) | -- | 6 M KOH Two electrode | 3 |
| **Seaweed derived carbon** | 3270 | 425 (0.1 A $g^{-1}$) | -- | 1 M $H_2SO_4$ Two electrode | 4 |
| **Hierarchical porous graphene** | 1533 | 280 (1 A $g^{-1}$) | 224 (1 A $g^{-1}$) | 6 M KOH Two electrode | 5 |
| **Polypyrrole-Derived activated carbon** | 3432 | 300 (1 mV $s^{-1}$) | -- | $EMMBF_4$ Two electrode | 6 |
| **Holey graphene frameworks** | 810 | 310 (1 A $g^{-1}$) | 220 (1 A $g^{-1}$) | 6 M KOH Two electrode | 7 |
| **Graphene** | -- | 200 (1 A $g^{-1}$) | 255 (0.1 A $g^{-1}$) | 1 M $H_2SO_4$ Two electrode | 8 |
| **Dead leaves derived carbons** | 1230 | 400 (0.5 A $g^{-1}$) | -- | 1 M $H_2SO_4$ Two electrode | 9 |
| **Reduced graphene oxide** | 2400 | 310 (20 A $g^{-1}$) | -- | 1 M KOH Two electrode | 10 |
| **Graphene-based porous frameworks** | 350 | 226 (1 mV $s^{-1}$) | -- | 1 M $H_2SO_4$ Two electrode | 11 |
| **Eggshell membranes derived carbon** | 221 | 297 (0.2 A $g^{-1}$) | -- | 1 M KOH Two electrode | 12 |
| **3D microporous graphene frameworks** | 194 | 202 (1 A $g^{-1}$) | -- | 1 M $Na_2SO_4$ Two electrode | 13 |
| **Natural organic chemicals derived carbons** | 2967 | 230 (0.1 A $g^{-1}$) | 100 (0.1 A $g^{-1}$) | 1 M $TEAFB_4$/AN Two electrode | 14 |
| **Activated graphene** | 3100 | 165 (1.4 A $g^{-1}$) | 60 (1.4 A $g^{-1}$) | 1 M | 15 |

| | | | | | |
|---|---|---|---|---|---|
| | | | | $BMIMBF_4$/AN<br>Two electrode | |
| **3D-printed graphene aerogel lattice** | 418 | 64 (10 A $g^{-1}$) | 2 (10 A $g^{-1}$) | 3 M KOH<br>Two electrode | 16 |
| **Carbon cryogel** | 1025 | 161 (10 A $g^{-1}$) | 80 (10 A $g^{-1}$) | 1 M $H_2SO_4$<br>Two electrode | 17 |
| **Nanoporous carbon** | 1472 | 220 (0.5 A $g^{-1}$) | -- | 6 M $H_2SO_4$<br>Two electrode | 18 |
| **KOH activated $C_{70}$ microstructure** | 1249 | 192 (5 A $g^{-1}$) | -- | 1 M $H_2SO_4$<br>Two electrode | 19 |
| **Superactivated carbon** | 1600 | 246 (0.2 A $g^{-1}$) | -- | 1 M $H_2SO_4$<br>Two electrode | 20 |
| **Graphene/carbon** | 492 | 250 (5 A $g^{-1}$) | -- | 1 M $Na_2SO_4$<br>Two electrode | 21 |
| **Hierarchically porous carbons** | 2988 | 238 (0.2 A $g^{-1}$) | -- | 6 M $KOH_4$<br>Two electrode | 22 |
| **Fungi derived porous carbon** | 80 | 196 (5 mV $s^{-1}$)) | 180 (5 mV $s^{-1}$)) | 6 M $KOH_4$<br>Two electrode | 23 |
| **Ordered mesoporous carbide-derived carbon** | 2250 | 170 (0.1 A $g^{-1}$) | 178 (0.1 A $g^{-1}$) | 1 M $TEAFB_4$/AN<br>Two electrode | 24 |
| **Dry elm samara** | 1947 | 470 (1 A g-1) | -- | 6 M KOH<br>Three electrode | 25 |
| **Protein** | 805.7 | 390.4 (0.2 A g-1) | -- | 1 M H2SO4<br>Three electrode | 26 |
| **Human hair** | 1306 | 340 (1 A g-1) | -- | 6 M KOH<br>Three electrode | 27 |
| **Fungus** | 1103 | 373 (0.5 A g-1) | -- | 6 M KOH<br>Three electrode | 28 |
| **Shiitake mushroom** | 2988 | 306 (1 A g-1) | -- | 6 M KOH<br>Three electrode | 29 |

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