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Bamboo-inspired cell-scale assembly for energy device applications

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Rapid advances in flexible and wearable smart textiles demand low-cost, high-energy/power-density fiber-shaped supercapacitors (FSCs). The performance of FSCs is determined by the fabrication and assembly of fiber-shaped electrodes (FSEs), where an active charge-storage material is always clad around flexible charge transmission current collectors. Inspired by the tissue structure of natural bamboo, wherein parenchyma cells (PCs) that store nutrients are clad around bamboo fibers (BFs), we propose a strategy for converting bamboo cells into FSEs using conductive BFs and activated PCs as current collectors and active materials, respectively. The assembled electrode has a high specific capacitance of 1454 mF cm^{-2} at 0.64 mA cm^{-2} . A solid-state FSC with a pair of bamboo-structured electrodes exhibited a substantially high energy density. Its mechanical flexibility enabled the knitting of wearable wristbands to drive ultra-small voltmeter indicators. This lightweight, low cost, and high-energy-density bamboo-structured FSC could enable numerous smart textile applications.

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INTRODUCTION

Flexible fiber/yarn-based supercapacitors (FSCs) are widely used as energy-storage devices for wearable electronics owing to their high capacity to be miniaturized and knitted into textiles with the required shape^{1–5}. Most FSCs consist of two opposite electrodes separated by a gel electrolyte. In this context, the key issue is to fabricate fiber-shaped electrodes (FSEs) and assemble them into a fiber-shaped device^{6,7}. FSEs can be realized by cladding activated materials around current collectors, that is, conductive fibers⁴. In achieving high capacitive performances, FSEs require high mass loadings of high-surface-area active materials and outstanding conductivity of fibrous scaffolds while maintaining mechanical flexibility^{8,9}. Various fibrous scaffolds have been employed, such as carbonaceous fibers^{1,10}, metal wires^{11,12}, synthetic polymer fibers^{8,13}, and natural fibers^{4,14,15}. Among them, natural fibers from biomass such as cotton have garnered substantial attention for the fabrication of FSEs because of their low cost, low weight, and natural sustainability^{4,16}.

Compared with other biomass materials, bamboo has special advantages, such as a high growth rate, high fiber strength and extraordinary large deflection^{17,18}. More importantly, its tissue structure is relatively simple and is mainly composed of parenchyma cells (PCs) and fiber cells (FCs)¹⁹. PCs, which contribute to nutrient storage and transport, are tightly clad around FCs which provide mechanical support and facilitate water and inorganic salt transport^{18–20}. The hollow ultra-high aspect ratio of FCs can be considered the natural long one-dimensional fibers with electron transfer potential. Note that there are plenty of nano-sized and micron-sized pores in the cell wall of PCs. These natural pore structures originally provide channels for biomass exchange between adjacent cells and also facilitate energy storage. These pores are capable of supplying extra access to the surrounding cells and thus shorten the distances for ionic diffusion²¹. Interestingly, this tissue structure and functionality are highly similar to those of FSEs in FSCs, wherein activated materials

with the charge-storage function surround current collectors with the electron transmission function¹⁹. Moreover, the moisture contained in bamboo is analogous to the electrolyte contained in FSCs. On the one hand, FCs are good scaffolds for aqueous electroless plating to realize conductivity owing to their strong affinity to water^{4,22}. Notably, delignified FCs retain the orientation of cellulose and form an interconnected porous structure with abundant microfibrils, turning into cellulosic bamboo fibers (BFs)²³. The multiscale network structure is susceptible to compressive, bending, stretching, and torsional deformations, which enable the fibers to adsorb conductive nanoparticles and exhibit excellent flexibility²⁴. This makes BFs highly suitable as current collectors in FSEs. On the other hand, PCs with a multiwall layer structure (favorable for etching to form pores) and a pit structure (beneficial for complete impregnation by the activator) are suitable as the precursor material for the active materials in FSEs^{20,24,25}. Thus, the natural bamboo structure can be reconstructed to form FSEs from the original bamboo cells after functionalization.

In this study, based on cell-scale design, we developed bamboo-structured FSCs using conductive Ag-coated BFs and activated PCs. Specifically, Ag nanoparticles (Ag NPs) were reduced in situ within the directional 3D interconnected porous structure of BFs, producing a highly conductive fiber substrate. With carbonization and activation, the high-surface-area PCs enabled the formation of a delicate pore structure and multiscale pore sizes. Furthermore, they possessed good compatibility with FSC devices and suitably large contact areas. Therefore, the bamboo-structured FSC achieved a high capacitance and energy density with good bending fatigue retention. Hence, these flexible and lightweight FSCs can be easily integrated in wearable electronic devices. Our success in the evolution of flexible FSCs from natural bamboo may not only open up the possibility of fabricating the sustainable high-performance wearable energy device, but also provide potential value for the structural and

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interface design and the high-value utilization based on bamboo cells.

RESULTS AND DISCUSSION

Fabrication of flexible bamboo-structured FSCs

The design concept and fabrication process of the flexible bamboo-structured FSC are shown in Fig. 1a. The structure of the FSC is very similar to that of natural bamboo, where PCs surround the BF (Fig. 1b, c). The next step is to functionalize the two kinds of cells respectively and then recombine them. First, we separated BFs and PCs, removing hemicellulose and lignin from the bamboo (Supplementary Fig. 1,2). The BFs were further electroless plated with Ag to provide electron conduction capability. Subsequently, the PCs were carbonized and activated using KOH to create numerous micropores and mesopores, which increased their charge-storage ability. The bamboo-structured FSEs were easily fabricated by reassembling the conductive BFs and activated PCs. Finally, two FSEs were coated with a thin layer of PVA-CMC-KOH gel electrolyte and then twisted together to form an all-solid-state FSC.

Characterizations of conductive BFs

The special fibrous structure of BFs can be used naturally or artificially to accommodate a wide range of applications^{21,26,27}. The micron-sized single FC embedded in vascular bundle can be anatomically perceived as a hollow long fiber composed of approximately 10 layers of walled structures with different microfibril angles of cellulose, which is embedded in an interwoven ligneous matrix. After removing the matrix from the cell wall, BFs retained the cellulose skeleton and exhibited a 3D interconnected porous structure (Fig. 2a and inset in Fig. 2b).

Accordingly, the porosity of BFs increased from 34.10 to 83.00% due to the presence of macropores and micropores (Fig. 2b). The fibrous and interconnected porous structure seems to be preferable for energy storage applications such as supercapacitors because it can provide large inner surface areas and enrich electrochemically active sites compared to solid counterpart²¹. On the other hand, the rich and uniform internal space provides a natural template for loading various functional particles. Therefore, we took advantage of the special structure of BFs to load Ag NPs to make them conductive. Supplementary Fig. 3 and Fig. 2c show the artificial-color scanning electron microscopy (SEM) images (Supplementary Method 1) of the BFs during electroless Ag plating. At the beginning of Ag plating, the pores on the fiber surface filled with Ag NPs and eventually formed a complete layer on the surface (Supplementary Fig. 4), as confirmed by the corresponding elemental maps (Fig. 2c). SEM analysis of a fiber cross-section revealed numerous homogeneously distributed Ag NPs (Fig. 2d). Therefore, the interconnected porous structure skeleton of BFs obtained by removing lignin and hemicellulose was appropriate for achieving a high loading of Ag NPs, quantified as 54.5 wt.% using ICP-MS. Abundant Ag NPs wrapped the BFs (Fig. 2e), and the Ag NPs were approximately spherical, with a particle size range of 1.3–40 nm after dispersing (Supplementary Fig. 5a, b). High-resolution transmission electron microscopy (HRTEM) images of the dispersed Ag NPs revealed a lattice fringe spacing of ~0.20 nm corresponding to the (111) plane of the face-centered-cubic Ag crystal (Fig. 2f)²⁸. The selected area electron diffraction (SAED) pattern was a ring composed of discontinuous bright spots, indicating highly crystalline Ag NPs.

Figure 2g shows the diffraction patterns of the BFs before and after Ag plating. The pristine BFs exhibited a broad peak at $2\theta = 15.8^\circ$ (110) and a single sharp peak at $2\theta = 22.2^\circ$ (200), corresponding to a typical cellulose I structure²⁹. After plating,

Fig. 1 Schematic of the design concept of flexible bamboo-structured FSCs and the tissue structure of natural bamboo. **a** Process for fabricating the flexible bamboo-structured FSCs. **(b–c)** SEM images of the natural bamboo: **b** top and cross-section of the natural bamboo, **c** cross-section showing the structural relationship between FCs and PCs in the natural bamboo.

Fig. 2 Structure and electrical conductivity of the Ag-coated BFs. **a** Diagram of cell wall structure of bamboo fibers and the hollow 3D interconnected porous structure after cellulose skeleton retaining process. **b** The incremental pore volume of pore distribution and cumulative pore volume of pore distribution of FCs and BFs; inset: artificial-color SEM images of the pristine BF. **c** Artificial-color SEM images of the conductive BF; inset: element maps of C, O, and Ag for the sample. **d** SEM images of the cross-section, magnified image and elemental maps of C, O, and Ag of an Ag-coated BF. **e** TEM image of conductive BF. **f** HRTEM image of an individual Ag NP after dispersion, showing the (111) lattice plane of Ag; inset: corresponding SAED image. **g** XRD patterns; inset: the local enlarged region ($2\theta = 8-15^\circ$). **h** XPS profiles, and **i** stress-strain curves of pristine and conductive BFs. **j** Conductivity of pristine BF (0 S m^{-1}), commercial CF (1000 S m^{-1}), and our conductive BF (43300 S m^{-1}); inset: a single conductive BF bundle used as a wire to light an LED.

the BFs presented a typical cellulose II structure with a characteristic peak in the region of $10-15^\circ$ (1-10) (the inset in Fig. 2g)³⁰. The characteristic diffraction peaks of Ag at 38.0° (111), 44.2° (200), 64.3° (220), and 77.2° (311) were clearly observed, indicating a high Ag NP loading on the BFs³¹. This was further proved by the X-ray photoelectron spectroscopy (XPS) peaks corresponding to zero-valent Ag at 374.1 eV ($\text{Ag}^0 3d_{3/2}$) and 368.1 eV ($\text{Ag}^0 3d_{5/2}$) (Fig. 2h)²⁸. Furthermore, in situ reduction and aggregation of Ag NPs within the BFs could fill the pores and enhance the mechanical performance of the microfibril network. Stress-strain tests showed that the Ag NP coating increased the tensile strength and elastic modulus of the conductive BFs by 42.1% and 38.9%, respectively, compared with those of the pristine BFs (Fig. 2i). The significant improvement in the strain of the conductive BFs was attributed to the transformation of the crystal structure from cellulose I to cellulose II owing to treatment with a highly concentrated alkaline solution. The conductivity of the Ag-coated BFs was $43,300 \text{ S m}^{-1}$ (Fig. 2j), which is much higher than that of commercial carbon fiber (CF; 1000 S m^{-1}), whose single fiber bundle could be used as a wire to light a light-emitting diode (LED). Even when the Ag NPs were coated with a low-concentration (8 times lower concentration) $\text{Ag}(\text{NH}_3)_2\text{NO}_3$ solution, a high conductivity of $19,600 \text{ S m}^{-1}$ was achieved (Supplementary Fig. 6). The high conductivity was attributed to the surface roughness of the BF and its interconnected porous structure, which were suitable for filling the Ag NPs. The final diameter of a single conductive BF bundle was 0.106 mm (Supplementary Fig. 7). A natural FC bundle was electroless Ag

plated by the same method. However, its conductivity was five orders of magnitude lower than that of Ag-coated BFs (Supplementary Fig. 8), fully illustrating the advantages of the interconnected porous structure. Notably, the aim of this strategy was to make the BFs conductive, and not necessarily to achieve a high loading of Ag NPs. Low-cost metals such as Cu and Ni could also be facilely and efficiently loaded onto the BFs via in situ autocatalytic electroless plating (Supplementary Figs. 9, 10). A single Cu-coated BF bundle and Ni-coated BF bundle could also be used as a wire to light an LED.

Characterizations of activated PCs

Active materials for SCs usually have high porosity for charge storage. PCs themselves have abundant natural pores with a diameter of $\sim 1.3 \mu\text{m}$ (Fig. 3a). We further increased the number of micropores in the PCs by a simple carbonization and activation method. The PCs, such as those from the pristine BFs, had a cellulose network structure formed by the removal of hemicellulose and lignin. After carbonization, the PCs shrunk, and the surface densified (Supplementary Fig. 11a, b). The Raman spectrum of the carbonized PCs showed both D and G peaks, with $I_D/I_G = 0.90$, indicating the dominance of the graphite structure (Supplementary Fig. 12). The structure of the PCs tended to be ordered during carbonization. After KOH activation, numerous nanoscale pores appeared on the surface (Fig. 3b), and the I_D/I_G ratio increased to 1.05, indicating the existence of a higher proportion of disordered domains after KOH activation

Fig. 3 Structure and porosity of activated PCs. Artificial-color SEM images of **a** pristine and **b** KOH-activated PC. **c** Raman spectra of the pristine and activated PC. **d** TEM (**d**₁), HRTEM image (**d**₂) and SAED pattern (**d**₃) of activated PC indicating the pore structure and amorphous structure. **e** Structural evolution of the PC from a parallel cellulose chain structure to the disordered graphite structure. **f** N₂ adsorption/desorption isotherms of pristine and activated PCs; inset: pore-size distribution curves of pristine and activated PCs. **g** XPS spectrum of pristine PCs and activated PCs and curve fitted high resolution XPS scans for C 1s and O 1s.

(Fig. 3c)³². The lack of diffractive points in the fast-Fourier-transform image and abundant disorder domains in the HRTEM image (Fig. 3d₃) further verified the formation of the amorphous structure.

The TEM image clearly showed mesopores with a diameter of ~5 nm (Fig. 3d₁), while the HRTEM images (Fig. 3d₂) showed abundant micropores. Hence, the KOH activation of the PCs resulted in a change from the parallel cellulose chain structure (Supplementary Fig. 13) to the disordered graphite structure (Fig. 3e). The N₂ adsorption/desorption isotherm of the activated PCs had a typical type-IV shape with an open knee and visible hysteresis, indicating abundant micropores and mesopores (Fig. 3f), while the pristine PCs had no distinct nanopores. After activation, the specific BET surface area (S_{BET}) increased by nearly 300 times (from 8.9 to 2523 m² g⁻¹). While the total pore volume (V_{tot}) of the pristine PCs was only 0.024 m³ g⁻¹, with dominant mesopores, V_{tot} increased to 1.554 m³ g⁻¹ after activation, with dominant micropores (Supplementary Fig. 14). According to the pore-size distribution (the inset in Fig. 3f), the pristine PCs had some mesopores and macropores (40–70 nm) from the cellulose network structure. In addition to numerous micropores (0.6–1.2 nm), the activated PCs also had abundant mesopores (2–50 nm). The small micropores help increase the capacitance of the SC by reducing the distance between the ion centers³³, while

the mesopores facilitate fast ion transport and increase the power/energy density^{32,34}. Figure 3g shows the full-range XPS spectra of pristine and activated PCs and the curve fitting for C 1s and O 1s. The carbonization and KOH activation are both a process of carbon enrichment with the increasing peak intensity of graphite and oxygen reduction (Supplementary Fig. 15). The as-formed surface oxygen-containing functional groups can increase the wettability of carbon materials and their affinity to electrolytes, and improve the ability of carbon materials to form electric double layers and capacitance characteristics³².

The electrochemical performance of the activated PCs was also evaluated in a three-electrode cell (Supplementary Method 2), and the quasi-rectangular CV curves and symmetrical triangular GC curves demonstrated outstanding electrochemical performance (Supplementary Figs. 16, 17). The PCs exhibited a high specific capacitance of 236 F g⁻¹ at 5 mV s⁻¹, which is much higher than that of many reported biomass carbon materials (Supplementary Table 1). PCs are good bio-based precursors to prepare functional carbon materials for electrocapacitive energy-storage applications owing to their original morphology, which has abundant hierarchical pores. Moreover, other activation methods, such as the addition of pseudocapacitive materials, could be adopted in future work to optimize the pore structure for other potential functional applications.

Ultra-high performance of bamboo-structured FSCs

To mimic the natural bamboo structure, we surrounded the conductive BF with the activated PCs (Supplementary Fig. 18a, b) to obtain a bamboo-structured FSE. The cycle washability showed the firmness of the Ag NPs layers and activated PCs (Supplementary Method 3 and Supplementary Fig. 19). Figure 4a shows an extended depth-of-field 3D image of the electrolyte-coated FSE (diameter = 0.93 mm). Supplementary Fig. 20 presents a schematic of the fiber structure and the interface between the activated PCs and conductive BFs. The nanoscale rough surface of the conductive BF contributes to improving the contact area, resulting in accessible amounts of activated PCs on the current collectors via electrostatic adsorption. A 1 mm-long FSE was

analyzed through nano-CT (Fig. 4b), whereby the obtained distributions of C and Ag are shown in Supplementary Fig. 21. Notably, the Ag NPs formed a penetrating network inside the electrode, facilitating electronic transfer through the electrode. The electrolyte coating was ~0.3 mm thick, and the FSE with twisted fibers was uniform in diameter (0.68 mm) and approximately circular in cross-section (Fig. 4c). The rough surface of the BFs and their internal cavity facilitated the adhesion of the PCs to the surface and within the fibers (Supplementary Fig. 22 and Fig. 4d). The Ag NPs on the fiber surface greatly increased the contact area between the PCs and the BFs (Supplementary Fig. 20), which reduced the internal resistance of the electrode. The conductivity of the FSE was $29,238 \text{ S m}^{-1}$, as measured by a resistance tester. The equivalent series resistance of a 6.5 cm-long FSE was

Fig. 4 Structural design of FSE and yarn FSC and their performance. **a** Extended depth-of-field 3D image of an electrolyte-coated FSE with (inset) a diameter of 0.93 mm. **b** 3D nano-CT image of a 1-mm-long FSE. **c** Extended depth-of-field 3D image of a FSE without an electrolyte coating (cross-section). **d** Artificial-color SEM images of the rough surface and internal cavity of the BF enabling good adhesion of the PCs in a FSE. Electrochemical performance of FSCs: **(e)** CV curves at various scan rates; **(f)** GC profiles at different current densities; **(g)** specific capacitance of FSC at different current densities; **(h)** Cycle performance of FSC with 91.6% capacitance retention after 8000 charge-discharge cycles at 7.49 mA cm^{-2} using a two-electrode configuration; Inset: two 6.5-cm-long FSCs connected in series to power an LED indicator. **i** Energy density and power density of FSC compared with published results: MnO_2/AC fibers ($36.0 \text{ } \mu\text{Wh cm}^{-2}$)⁴⁰, $\text{RGO}/\text{CNT}/\text{N-doped carbon fibers}$ ($11.2 \text{ } \mu\text{Wh cm}^{-2}$)³⁵, $\text{Ni wire}/\text{NiCo}_2\text{O}_4/\text{MnO}_2$ ($7.0 \text{ } \mu\text{Wh cm}^{-2}$)³⁶, $\text{Ni wire}/\text{CuCo}_2\text{S}_4$ fibers ($7.2 \text{ } \mu\text{Wh cm}^{-2}$)¹², highly activated carbon fibers ($26.0 \text{ } \mu\text{Wh cm}^{-2}$)⁴¹, $\text{PET}/\text{Au}/\text{Ni-MOF}/\text{carbon yarn}$ ($5.41 \text{ } \mu\text{Wh cm}^{-2}$)⁴², $\text{CNT}/\text{ordered mesoporous carbon}$ ($1.77 \text{ } \mu\text{Wh cm}^{-2}$)⁴³, $\text{CNTs}/\text{PEDOT:PSS}$ ($3.6 \text{ } \mu\text{Wh cm}^{-2}$)⁴⁴, $\text{CNFs}/\text{RGO}/\text{PPy}$ ($14.37 \text{ } \mu\text{Wh cm}^{-2}$)⁴⁵, $\text{RGO}/\text{MoS}_2/\text{P}$ ($1.44 \text{ } \mu\text{Wh cm}^{-2}$)⁴⁶; **j** Capacitance retention of a 6.5 cm-long FSC under various mechanical deformation conditions; inset: capacitance retention after 1000 bending (360°) cycles. **k** Photograph of our wearable wristband and the potential applications of such wearable devices, like smart bracelets.

$2.9 \Omega \text{ cm}^{-2}$ using a three-electrode configuration (Supplementary Fig. 23). It also exhibited high areal and linear capacitances of 1131 mF cm^{-2} and 72 mF cm^{-1} at 20 mV s^{-1} , respectively (Supplementary Fig. 24).

Finally, we constructed bamboo-structured FSCs by twisting two electrolyte-coated electrodes together and coating them with another thin layer of electrolyte. Supplementary Fig. 25 shows an extended depth-of-field 3D image of the cross-section of the bamboo-structured FSCs. The electrochemical performance of a 6.5 cm-long FSC was evaluated via CV and GC curves. The quasi-rectangular CV curves (Fig. 4e) and linear symmetric GC curves (Fig. 4f) indicated a high electrochemical double layer charging capacitance. Even at a high scanning rate (20 mV s^{-1}), the CV curve exhibited a slightly sloped rectangular shape. The highest values of the areal, volumetric, and linear capacitances of the FSE were 1454 mF cm^{-2} , 86 F cm^{-3} , and 310 mF cm^{-1} , respectively, calculated from the GC curve at a current density of 0.64 mA cm^{-2} . We also tested the capacitance of FSE made from nature bamboo fiber for comparison and it showed an extremely low areal, volumetric and linear capacitances of 0.17 mF cm^{-2} , 0.01 F cm^{-3} , and 0.04 mF cm^{-1} , respectively (Supplementary Fig. 26). On the one hand, this is due to the poor conductivity of the collector (Supplementary Fig. 8). On the other hand, the fewer Ag NPs and lower interface roughness led to poor contact between the collector and the active PCs, resulting the extremely high contact resistance (Supplementary Fig. 27). The results proved that the 3D interconnected porous structure of BF was indeed one of the key factors for the preparation of high-performance FSCs.

The performance of the FSE was superior to that of the previously reported FSEs, even those based on pseudocapacitive materials (Supplementary Table 2), such as GCF@NC@NCO fibers ($11.2 \mu\text{W h cm}^{-2}$)³⁵, Ni wire/NiCo₂O₄/MnO₂ ($7.0 \mu\text{W h cm}^{-2}$)³⁶, and Ni wire/CuCo₂S₄ fibers ($7.2 \mu\text{W h cm}^{-2}$)¹². Even at high current density (7.49 mA cm^{-2}), the capacitance of this device can maintain about 70%, showing good rate performance, which could be ascribed to the special structure of activated PCs and good electrode system³⁷. The porous structure promotes the good permeation of the electrolyte into the surface of electrode material, which directly decrease the diffusion distance of OH⁻ ions³⁸. The equivalent series resistance of a 6.5 cm-long FSC was $2.3 \Omega \text{ cm}^{-2}$ (Supplementary Fig. 28), which is much lower than that reported for a bisrolled FSC³⁹. This could be due to the high conductivity of the conductive BF and the good contact between the PCs and BF. This device showed a good cycling performance, with 91.6% capacitance retention after 8000 charge–discharge cycles at 7.49 mA cm^{-2} (Fig. 4h).

The areal energy density of the FSC was $65 \mu\text{W h cm}^{-2}$ at an areal power density of $119 \mu\text{W cm}^{-2}$, which is higher than that previously reported for FSCs (Fig. 4i), including those based on MnO₂@AC fibers ($36.0 \mu\text{W h cm}^{-2}$)⁴⁰, highly activated carbon fibers ($26.0 \mu\text{W h cm}^{-2}$)⁴¹, PET/Au/Ni-MOF@carbon yarn ($5.41 \mu\text{W h cm}^{-2}$)⁴², CNT@ordered mesoporous carbon ($1.77 \mu\text{W h cm}^{-2}$)⁴³, CNTs/PEDOT:PSS ($3.6 \mu\text{W h cm}^{-2}$)⁴⁴, CNFs/RGO@PPy ($14.37 \mu\text{W h cm}^{-2}$)⁴⁵ and RGO@MoS₂@P ($1.44 \mu\text{W h cm}^{-2}$)⁴⁶.

Figure 4j shows that this device maintained good performance when bent at arbitrary angles because of its high flexibility. Even after bending, it retained 93% of its initial capacitance, except when crumpled. A 92.4% retention rate was observed after 1000 bending (360°) cycles. The outstanding flexibility could be attributed to the synergistic advantages of BF and PC, which inherited the extraordinary deformation resistance of the natural bamboo. During increasing bending deflection, the deformation of PCs could facilitate the bending deformability, which were squeezed between the fiber bundles⁴⁷. The inset in Fig. 4h shows two 6.5 cm-long FSCs connected in series to power an LED. Owing to its outstanding mechanical flexibility, one 10 cm-long and five 20 cm-long FSCs were knitted together to form a wearable wristband (Supplementary Fig. 29), which was able to power an

ultra-small blue digital voltmeter. Such wearable devices could be applied in the development of an extensive range of smart devices, such as bracelets (Fig. 4k).

We successfully fabricated flexible bamboo-structured FSCs using BF and PC extracted from natural bamboo, which were first modified to increase their conductivity and porosity, respectively. The conductive BF was then wrapped with the activated PC to produce electrodes, which were then twisted together and coated with a thin layer of gel electrolyte to form an all-solid-state FSC. The electrodes had the highest areal capacitance reported to date for fiber/yarn electrodes. Similarly, the FSC exhibited a high energy density and outstanding mechanical flexibility with a low capacitance loss upon bending at arbitrary angles. We demonstrated the practical feasibility of an energy-storage device based on the FSCs for wearable electronics by fabricating a wearable wristband. Considering the long-term stability issue of PVA-KOH, we will try new methods for conducting cellulosic bamboo fibers and different gel systems in future work to improve the long-term stability of FSC. Our bamboo-inspired work presents a promising direction for developing energy-storage devices by using the BF and PC based on their biological structures. Examples of biological structures include the 3D interconnected porous structure of BF and the multiwall layer structure and pit structure of PC, which can be transformed into multifunctional and responsive materials.

METHODS

Materials and chemicals

Four-year-old moso bamboo (*Phyllostachy edulis*) stems were collected from a forest in Anji, Zhejiang, China. Acetic acid (CH₃COOH; AR; 99.5%), anhydrous sodium carbonate (Na₂CO₃; AR; 99.5%), potassium sodium tartrate tetrahydrate (C₄H₄O₆KNa·4H₂O; AR; 99%), boric acid (H₃BO₃; AR; 99.5%), ammonium chloride (NH₄Cl; AR; 99.5%), sodium hypophosphite monohydrate (NaH₂PO₂·H₂O; AR), hydrochloric acid (HCl; AR), and ammonium hydroxide (NH₄OH; AR; 25%) were purchased from Beijing Chemical Works. Formaldehyde solution (CH₂O; AR; 37%), copper sulfate pentahydrate (CuSO₄·5H₂O; AR; 99%), nickel(II) sulfate hexahydrate (NiSO₄·6H₂O; AR; 98.5%), palladium chloride (PdCl₂; 59–60%), citric acid trisodium salt (C₆H₅O₇Na₃; AR; 98%), and potassium hydroxide (KOH; AR; 90%) were purchased from Macklin. Sodium chlorite (NaClO₂; 80%), sodium hydroxide (NaOH; 97%), silver nitrate (AgNO₃; AR; 99.8%), glucose anhydrous (C₆H₁₂O₆; AR), and polyvinyl alcohol (PVA; alcoholysis degree 87.0–89.0%) were purchased from Aladdin.

Separation of BF and PC

The BF and PC were obtained by removing the lignin and hemicellulose from the bamboo cell walls, followed by freeze drying and shaking separation. The separation method used in this study is commonly used for the delignification of biomass materials, such as wood^{48,49}. Specifically, bamboo slices with dimensions of 200 mm (length) × 10 mm (width) × 0.5 mm (thickness) were immersed in 0.2 M NaClO₂ solution buffered with CH₃COOH at pH 4.5 and 80 °C in a water bath. The delignified slices were further treated with 2 M NaOH solution at 80 °C to remove the hemicellulose. Finally, the samples were washed with deionized water until they were pH-neutral and then freeze-dried at −30 °C for 72 h under vacuum (25 Pa). Finally, the BF and PC were collected by manual shaking.

Preparation of conductive BF

Conductive BF was prepared by autocatalytic electroless silver plating. Briefly, 2 wt% NH₄OH solution was added dropwise into an aqueous 2 M AgNO₃ solution to prepare a Ag(NH₃)₂NO₃ solution. After immersing the BF in 5 M NaOH solution at room temperature for 2 h, they were dried in an oven at 40 °C and immersed in a mixture of the Ag(NH₃)₂NO₃ solution and 1.7 wt% C₆H₁₂O₆ solution to react for 15 min.

To demonstrate the applicability of the design concept of bamboo-structured FSCs, conductive BF with a low-cost metal coating, including Cu and Ni coating, were prepared (Supplementary Method 4).

Preparation of activated PCs

The PCs were carbonized by heating in N_2 at 500 °C for 1 h. The activated PC was obtained by activating the carbonized cells with KOH (mass ratio of KOH/cell: 4:1) in N_2 at 800 °C for 1 h.

Preparation of bamboo-structured FSEs

To ensure the adherence of the activated PCs to the surface of the conductive BFs, the mass loading was increased by a repeated dipping-drying process. First, the PCs were dispersed in 100 mL ethanol (10 mg mL^{-1}) by ultrasonic dispersion at 25 °C for 30 min. The conductive BFs were then immersed in the PC suspension for 30 min. Subsequently, the fibers were dried in a vacuum-drying oven for 1 h. The mass loading of the activated PCs on the conductive BFs after the dipping-drying process was approximately 0.78 mg cm^{-1} . The density of the conductive BFs was controlled at 3 mg cm^{-1} . After drop-casting the dispersion on the loose conductive BFs at a total mass loading of 2.2 mg cm^{-1} , the hybrid fibers were twisted to 560 turns per meter using a handheld fiber spinner to fabricate the flexible bamboo-structured FSE, resulting in a tightened diameter of $\sim 0.68 \text{ mm}$ (without an electrolyte).

Bamboo-structured FSC assembly

The PVA-CMC-KOH gel electrolyte was prepared by heating a mixture of 3 g PVA, 1 g CMC, and 6 g of 10 M KOH in 60 ml deionized water at 85 °C for several hours until clear. Two FSEs were separately coated with a thin layer of PVA-CMC-KOH gel electrolyte via dip coating. After the gel solidified, the two FSEs were twisted together using a handheld fiber spinner and then coated with a thin layer of gel electrolyte to prepare bamboo-structured FSCs.

Characterization

The morphology and structures of the samples were characterized by SEM (Hitachi SU8020) coupled with an energy dispersive X-ray spectroscopy (EDS) (Horiba 7021-H) system; TEM (JEM-1200EX); high-resolution TEM (HRTEM; FEI Tecnai G2 F20); extended depth-of-field 3D microscopy (Keyence VHX-6000); high-resolution nano-computed tomography (nano-CT; Bruker, SkyScan 2211); X-ray diffraction (XRD; Bruker D8 Advance); and micro Raman spectrometry (PerkinElmer RamanStation 400).

The chemical structure and surface functional groups were characterized by Fourier transform infrared (FTIR; Nicolet IS10) spectrometry and XPS (Escalab 250Xi). The Ag content of the conductive BFs was determined using inductively coupled plasma mass spectrometry (ICP-MS). The porosity was determined by the physical adsorption of N_2 at -196°C using an Autosorb-iQ analyzer.

The mechanical properties of a single BF were measured using a custom-built fiber-gripping system combined with a small commercial high-resolution mechanical tester (Instron Microtester 5848). The electrical resistivity of the conductive BF was obtained based on a four-probe electrical resistivity measuring technique using a resistivity tester (YAOS FM100GH). The electrical resistivity of the FSE was measured by using a Keithley 2400 Source Meter. Electrochemical measurements of the bamboo-structured FSCs were performed using a two-electrode configuration and an electrochemical analyzer (CH Instrument CHI 760). The electrochemical measurements of the bamboo-structured FSE are shown in Supplementary Method 5.

Calculation of the electrochemical performance

The flexible bamboo-structured FSC is a symmetric two-electrode SC. The specific linear (C_{sl}), areal (C_{sa}), and volumetric capacitance (C_{sv}) values of the electrode in a two-electrode cell were calculated from galvanostatic charge-discharge (GC) profiles using the following equations:

$$C_{sl} = \frac{2 \times I \times \Delta t}{L_s \times \Delta V} \quad (1)$$

$$C_{sa} = \frac{2 \times I \times \Delta t}{A_s \times \Delta V} \quad (2)$$

$$C_{sv} = \frac{2 \times I \times \Delta t}{V_s \times \Delta V} \quad (3)$$

where I is the discharge current and $\frac{\Delta V}{\Delta t}$ is the slope of the discharge curve. In addition, L_s , A_s , and V_s are the total length, external surface area, and volume of a single bamboo-structured FSE, respectively. The capacitance

(C), areal density (E_a), and power density (P_a) of the SC were obtained from the following equations:

$$C = \frac{I \times \Delta t}{\Delta V} \quad (4)$$

$$E_a = \frac{C \times \Delta V^2}{2 \times A_{sc}} \quad (5)$$

$$P_a = \frac{E_a}{\Delta t} \quad (6)$$

Here, Δt and ΔV are the discharge time and potential difference, respectively, of an individual discharge cycle, and A_{sc} was assumed to be double the external surface area of an electrolyte-coated FSE.

DATA AVAILABILITY

All data needed to evaluate the conclusions in the paper are present in the paper and/or the Supplementary Materials. Additional data related to this paper are available from the corresponding author upon reasonable request.

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AUTHOR CONTRIBUTIONS

Q.L. and R.G. contributed equally to this work. Y.H. conceived the idea for this study and created the 3D illustrations. Q.L. contributed to material synthesis and most material characterizations. R.G. and Y.L. contributed to the electrochemical data collection. Q.L., R.G., Y.L., and Y.H. analyzed the data and wrote the manuscript. S.L. assisted in the data analysis and production of the figures. D.L., Y.Y., and W.Y. had scientific discussions and improved the manuscript. All authors reviewed and commented on the final manuscript.

COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

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