Fabrication of Robust, Highly Conductive, and Elastic Hybrid Carbon Foam Platform for High-Performance Compressible Asymmetry Supercapacitors

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ABSTRACT: Highly conductive and elastic three-dimensional (3D) porous carbon materials are ideal platforms to fabricate electrodes for high-performance compressible supercapacitors. Herein, a robust, highly conductive, and elastic carbon foam (CF) hybrid material is reported, which is fabricated by integrating cellulose nanofiber/multiwalled carbon nanotube (CNF/MWCNT) aerogel sheets with a melamine sponge (MS), followed by carbonization. The carbonized CNF/MWCNT aerogel sheets contribute to the high conductivity and specific surface area of the CF, and the 3D network-like skeleton derived from the carbonization of the MS enhances the elasticity and stability of the CF. More importantly, the CF possesses good scalability, allowing the introduction of electroactive materials such as polypyrrole (PPy) and Fe₃O₄ to fabricate high-performance compressible PPy−CF and Fe₃O₄−CF electrodes. Moreover, an assembled PPy−CF//Fe₃O₄−CF device shows reversible charging−discharging at a voltage of 1.6 V and demonstrates a high specific capacitance (172.5 F/g) and an outstanding energy density (59.9 W h/kg). The device exhibits capacitance retention rates reaching 98.3% and stable energy storage characteristics even under different degrees of compressive deformation. This study offers a scalable strategy for fabricating high-performance compressible supercapacitors, thereby providing a new means of satisfying the energy storage needs of portable electronic devices that are prone to deformation.

1. INTRODUCTION

With the rapid development of portable and wearable electronic devices, novel energy storage devices that can be subjected to deformations such as bending, compression, and tension have received increased attention.¹−⁴ Among the available options, supercapacitors are considered as optimal power supply units for portable and wearable electronic devices because they offer outstanding power densities, long cycle lives, and higher charge and discharge rates than those of batteries.⁵−⁹ In recent years, numerous novel high-performance supercapacitors¹⁰−¹⁴ have been developed to satisfy the requirements of future portable and wearable electronic devices. Compressible supercapacitors¹⁵−¹⁸ are light, highly elastic, and fatigue resistant. Furthermore, their electrochemical behavior remains stable even in working environments involving considerable compressive deformation, allowing them to satisfy the energy needs of portable electronic products. These features have driven research on compressible supercapacitors. However, unlike the assembly of traditional supercapacitors, that of high-performance compressible supercapacitors requires an electrode material with good compressibility¹⁰,²⁰ in addition to excellent capacitive properties, conductivity, and stability.²¹−²³

Among assorted high-performance electrode materials, three-dimensional (3D) porous materials²⁴−²⁸ with unique nanostructures are ideal candidates for the construction of high-performance flexible and/or compressible supercapacitor electrodes. With hierarchical porous structures ranging from the nanometer to the millimeter scale and excellent mechanical flexibility and compressibility, highly conductive sponge-like materials²⁹,³⁰ show suitability for use in multiple types of high-performance energy storage devices, especially in compressible supercapacitors. Presently, three types of sponge-like materials can be used in high-performance compressible electrodes: (1) 3D porous carbon-based materials or hybrids thereof, prepared from highly conductive graphitic carbon materials such as graphene and carbon nanotubes;³¹−³⁴ (2) carbon materials or hybrids thereof derived from polymeric sponges;³⁵,³⁶,¹⁷,¹⁸,³⁷,³⁸,³⁹,⁴⁰ (3) and metal foams and derivatives thereof.⁴¹ In particular,
2. EXPERIMENTAL SECTION

2.1. Materials and Chemicals. Commercial MS was purchased from Sichuan Chemical Works Group. Bamboo CNFs (0.8 wt %) were provided by the International Center for Bamboo and Rattan (Beijing). MWCNTs (TNIM2, 8–15 nm in diameter) were purchased from Chengdu Institute of Organic Chemistry. Pyrrole monomer, ferric chloride (FeCl₃·6H₂O), polyvinyl alcohol (PVA), ethylene glycol, sodium acetate, ethylene diamine, lithium chloride (LiCl), and ethanol were obtained from the Aladdin Industrial Corporation. All reagents were of analytical grade and were used without further purification. Deionized water was used in all experiments.

2.2. Preparation of the CF Platform and Control Samples. The typical preparation process was modified according to our previous work. First, the commercial MS was pretreated with ethanol and deionized water three times and then vacuum-dried at 60 °C. Subsequently, the dried MS was immersed in a 20 mL mixed solution of HCl and FeCl₃ with a mass ratio of 1.5:1. The dispersion was evenly distributed in the pore structure of the sponge by repeatedly squeezing the MS. The filled MS was then placed in a freeze dryer at −50 °C for vacuum drying at 4.5 Pa for 4 days. This soaking and freeze-drying cycle was repeated three times. Finally, the obtained material was placed in a carbonization furnace, heated to 600 °C for 1 h at a rate of 2 °C/min under a mixed atmosphere (N₂/CO₂ = 1:1), and then continuously heated to 800 °C for 2 h at a rate of 3 °C/min to obtain the CF hybrid material with high specific surface area.

To demonstrate the advantages of CF as a platform for preparation of high-performance compressible electrodes, carbonized MS derivatives without MWCNTs and CNFs, but prepared under otherwise equal conditions, were used as reference samples, denoted as MS derivatives. Moreover, CNF/MWCNT carbon aerogels prepared under the same conditions were used as a control sample, denoted as CAs.

2.3. Preparation of PPY−CF and Fe₃O₄−CF Compressible Electrodes. The preparation of the PPY−CF electrode was as follows. First, 40 μL of the pyrrole monomer and 10 mL of ethanol were transferred by a pipette into a 50 mL flask under a N₂ atmosphere and stirred well. Thereafter, a CF hybrid material measuring 4 cm × 2 cm × 0.5 cm was completely immersed in the above mixed solution. It was placed in a 20 mL mixed solution of HCl and FeCl₃ with a concentration of 0.7 mol/L and then in a 2 °C water bath to continue polymerization for 6 h. Subsequently, the sample was sequentially cleaned with 0.3 mol/L HCl, 0.1 mol/L NaCl solution, and deionized water. The washing process was repeated five times. Finally, the sample was placed in a freeze dryer at −50 °C for 24 h and then vacuum-dried at 4.5 Pa for 3 days to obtain the compressible PPY−CF positive electrode.
photoelectron spectroscopy (XPS) tests were performed using powder X-ray diffraction (XRD) patterns were obtained using a Thermo ESCALAB 250XI. The compressive stress–strain curves were obtained using a WDW-100 tester at the compression and release rate of 2 mm/min, with all the samples measuring 2.0 cm × 2.0 cm × 1.1 cm. Atomic absorption spectrometry (ICE3000, Thermo Fisher Scientific) was performed to determine the Fe3O4 load in the electrode. Because the CF contains N element, the loading amount of PPy is determined by weighing on a physical balance. N2 adsorption was measured using an ASAP 2020 surface area and pore analyzer (Micromeritics instrument) at 77 K; all samples were degassed at 200 °C under vacuum for 8 h.

2.6. Electrochemical Measurements. An electrochemical workstation (CHI 660E) was used to perform electrochemical tests of the electrodes and assembled device. The electrochemical tests of the PPy–CF and Fe3O4–CF compressible electrodes were performed using a three-electrode system with 5 M LiCl as the electrolyte, a Pt plate as the counter electrode, and a saturated calomel electrode as the reference electrode. The working electrode had an effective area of 1.0 cm × 1.0 cm. The electrochemical tests of the devices were performed using a two-electrode system on the electrochemical workstation, and the effective area of the PPy–CF//Fe3O4–CF-ASC device was 1.0 cm².

3. RESULTS AND DISCUSSION

3.1. Preparation and Characterization of CF Platform, PPy–CF and Fe3O4–CF Electrodes. As illustrated in Figure 1a, the PPy–CF and Fe3O4–CF compressible electrodes are fabricated with the CF as the base through in situ polymerization and hydrothermal reactions. The typical process is as follows. First, the CNF/MWCNTs hybrid aerogel sheets are stacked onto the 3D network-like framework of the MS through the self-aggregation of MWCNTs and CNFs during the freeze-drying process. This yields a CNF/MWCNT/MS hybrid material with a 3D porous structure. Subsequently, high-temperature carbonization is performed to obtain a compressible, highly conductive, and robust CF platform. As shown in Figure S1, the CF is a porous hybrid...
material with a more ingenious structure formed by attaching several aerogel sheets to its continuous 3D network-like skeleton. On the one hand, compared with the 3D narrow skeleton structure of the MS derivatives (Figure 1b,c), the large and continuous carbon aerogel sheets (Figure 1d,e) derived from carbonizing the CNF/MWCNT (inset of e) sheets provide a higher external surface area and conductivity, which are advantageous for the subsequent loading of electroactive materials. On the other hand, the stress–strain curve (Figure S2) indicates that the CF has better compressibility and stability than the CNF/MWCNT CAs; this is mainly attributed to the 3D framework derived from carbonizing the MS (Figure 1b,c) that has a reinforcing effect on the CF, while the CAs are formed by accumulation of disordered aerogel sheets (Figure S3). Therefore, the CF possesses good compressibility, high surface area, and excellent conductivity, making it a suitable platform for the introduction of electroactive materials.

To further elucidate the advantages of the CF in terms of its microstructure, the porous structures of the MS derivatives and CF were also probed using N₂ adsorption. As shown in Figure S4a, the MS derivatives and CF exhibit a combination of type I and type IV adsorption–desorption isotherm curves, indicating the presence of micropores and mesopores. It should be noted that the CF shows more obvious microporous adsorption–desorption characteristics than MS derivative at the low relative pressure region ($P/P_0 = 0.05$), which indicates the presence of a lot of micropores. This is because the uniformly dispersed MWCNT and CNF in MWCNT/CNF hybrid aerogel sheets resulted in a large number of slit-like micropores (Figure 1e). Moreover, the corresponding pore size distribution (PSD) curve (Figure S4b) further confirms that the CF and MS derivatives have microporous and mesoporous structures. The 3D continuous macropores formed by the aerogel sheets are not reflected in the PSD curve but are verified using SEM (Figures 1d and S1). Furthermore, as summarized in Table S1, the CF possesses a Brunner–Emmet–Teller (BET) specific surface area of 86.7 m$^2$/g, which is much higher than that of the MS derivatives (114.1 m$^2$/g). Meanwhile, the CF also benefits from the structural stability of the CNF/MWCNT aerogel sheet during high-temperature carbonization and its protective effect on the MF skeleton. Therefore, the size of the CF hybrid aerogel sheet remains almost unchanged after high-temperature carbonization (Figure S5). In addition, this unique hybrid composition and structure also ensure that the CF is lightweight (Figure S6). Lastly, in situ embedding of PPy and Fe$_3$O$_4$ nanoparticles via polymerization and solvothermal reaction was conducted to prepare the PPy–CF and Fe$_3$O$_4$–CF compressible electrodes. During the polymerization and solvothermal reactions, the size and morphology of the CF platform remained unchanged (Figure S7a–c), indicating its good structural stability. This stability ensured successful subsequent assembling of the high-performance compressible ASC.

As next, we used SEM to evaluate the suitability of the CF as a platform for the introduction of electroactive materials by examining the microstructures of the PPy–CF and Fe$_3$O$_4$–CF electrodes. Figures S7b and 2a show the photograph and SEM image of the PPy–CF electrode material prepared using the CF as a platform, respectively. After polymerization, the PPy–CF electrode material retained the same bulk shape as that of the CF platform and its continuous 3D porous structure, which was attributed to the good structural stability and mechanical strength of the CF platform. Furthermore, the high-magnification SEM images (inset of Figures 1e and 2b) show that the MWCNT surfaces of the PPy–CF electrode are significantly coarser, possibly because they are loaded with PPy nanoparticles. To further determine the PPy loading in the PPy–CF electrode, we performed characterization using EDX spectroscopy. The EDX spectroscopy mapping images (Figure 2e) show uniform distributions of C and N in the PPy–CF electrode, indicating that PPy is successfully and uniformly loaded in situ onto the CF platform through polymerization.

To further demonstrate the versatility of the CF in the fabrication of high-performance electrode materials, we prepared a Fe$_3$O$_4$–CF electrode through a hydrothermal reaction. As shown in Figures S7c and 2c, the Fe$_3$O$_4$–CF electrode material retains its morphology and continuous 3D porosity after the hydrothermal reaction, providing further

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**Figure 2.** SEM images of (a) PPy–CF, (c) Fe$_3$O$_4$–CF, and corresponding high-magnification SEM images (b,d); (e) SEM image of PPy–CF and corresponding energy-dispersive X-ray (EDX) mapping images showing elemental C, N, and O; and (f) SEM images of Fe$_3$O$_4$–CF and corresponding EDX mapping images of elemental C, Fe, and O.
proof of the good structural stability and mechanical strength of the CF. The high-magnification SEM image of the Fe₃O₄−CF electrode material (Figure 2d) shows that the electroactive Fe₃O₄ nanoparticles (~500 nm) are successfully loaded onto the aerogel sheets through the hydrothermal reaction. Furthermore, the EDX spectroscopy mapping image of the corresponding Fe and O elements is displayed in Figure 2f. Elemental Fe and O are uniformly distributed over the corresponding selected regions, indicating that the Fe₃O₄ nanoparticles are uniformly attached in the Fe₃O₄−CF electrode. In addition, the magnetic adsorption experiment result, as shown in Figure S8, proves that the Fe₃O₄ is successfully introduced into the Fe₃O₄−CF electrode. This result further indicates that the CF possesses excellent versatility as a platform for introducing electroactive materials.

Although the CF platform has good structural stability, the in situ polymerization, hydrothermal reaction, and post-treatment (such as multiple washings with different solutions) processes may affect its microstructure. As a result, the 3D continuous pores of the PPy−CF and Fe₃O₄−CF electrodes were obviously not as dense and uniform as those of the CF. This may be because some of the CNF/MWCNT-derived carbon aerogel sheets fell off the MS skeleton during subsequent processing.

Furthermore, XPS and XRD analyses were conducted to probe the surface elemental valence states and chemical compositions of the CF, PPy−CF, and Fe₃O₄−CF electrodes. As shown in the C 1s spectrum in Figure 3a, the main peak at 284.6 eV corresponds to the characteristic graphitic carbon (C−C) peak of the MWCNT and the shoulder peak at 285.2 eV corresponds to the characteristic C=N peak of the carbonized derivative of the MS. Figures S9a and 3b,c show the full XPS spectra and corresponding C 1s and N 1s spectra for the PPy−CF electrode. The curve-fitted C 1s spectrum exhibits four characteristic peaks at 283.4, 284.1, 284.8, and 288.3 eV. The peaks at 284.8 and 284.1 eV correspond to the α-carbon (C−N) and β-carbon (C−C) of pyrrole, respectively. The characteristic peak at 288.3 eV corresponds to the carbonyl group double bond (C=O) formed through corrosion by water molecules during the electrochemical polymerization process. The intensity of the C−N characteristic peak is significantly enhanced in the spectrum of the PPy−CF electrode than that observed in the spectrum of the CF, which further indicates the successful loading of PPy. In addition, the curve-fitted N 1s spectrum for PPy−CF shows three characteristic peaks originating from PPy, at 399.7, 400.4, and 402.1 eV. These peaks correspond to −NH−, −N+H, and = N+−, respectively, in the main chain of PPy. The above results indicate that PPy is successfully introduced into the PPy−CF electrode, which is consistent with the SEM and EDX results.

Figures S9b and 3d,e show the full XPS spectrum and the corresponding Fe 2p and O 1s spectra for the Fe₃O₄−CF electrode. The curve-fitted Fe 2p spectrum exhibits two characteristic peaks at 711 and 725 eV, which are consistent with the characteristic Fe 2p₃/2 and Fe 2p₁/2 peaks obtained for Fe₃O₄ reported in earlier studies. Therefore, we deduced that the electrode material contained Fe₃O₄. Figure 3e shows three characteristic peaks in the curve-fitted O 1s spectrum at 529, 530.2, and 531.8 eV, which correspond to the bond energy peaks of Fe−O, Fe−O−C, and C−O, respectively. This further proves that the electrode material comprises C, Fe, and O. In addition, the XRD patterns of Fe₃O₄−CF exhibit six characteristic diffraction peaks at 2θ = 30.2, 35.5, 43.2, 53.4, 57.3, and 62.8° (Figure 3f), indicating the successful loading of Fe₃O₄ onto the
Figure 4. (a) Compressive stress–strain curves of the CF at different strains of 30% (black), 50% (blue), and 70% (red); (b) digital photographs showing the CF used as a connecting wire lighting an LED during compression and release cycles (photograph courtesy of Xueqin Zhang, Copyright 2021); (c) photograph showing bulk resistance measurement of the CF platform with a multimeter (photograph courtesy of Xueqin Zhang, Copyright 2021); (d,e) SEM images of the CF after 10 cycles of 50% compression; and (f) compression stress–strain curves of the PPy–CF and Fe₃O₄–CF electrodes after 1, 5, and 10 compression cycles at 30% strain.

Figure 5. (a) CV curves of CF and PPy–CF electrodes at a scan rate of 100 mV/s; (b) CV curves of CF and Fe₃O₄–CF electrodes at a scan rate of 100 mV/s; (c) area-specific capacitance and capacitance retention rate as a function of current density; and (d) Nyquist impedance spectra of CF, PPy–CF, and Fe₃O₄–CF electrodes.
Fe$_3$O$_4$—CF electrode. Notably, the characteristic diffraction peaks of PPy at $2\theta = 25.8^\circ$ in the PPy—CF electrode$^{52,53}$ overlap with the (001) peak of carbon in the CF because of the low peak intensities of PPy.

3.2. Physical Properties of the CF Platform, PPy—CF and Fe$_3$O$_4$—CF Electrodes. Because the CF is used as a carrier for the electroactive materials, namely, PPy and Fe$_3$O$_4$, its mechanical properties directly influence the compressibility of the PPy—CF and Fe$_3$O$_4$—CF electrodes. Figure 4a shows the stress—strain curves of the CF after 1, 5, and 10 cycles under 30, 50, and 70% compression. All curves exhibit similar trends and consist of three regions with different characteristics,$^{32,33}$ namely, the elastic, stable, and densification regions. This shows that the CF has excellent compressibility and can be used as an electroactive material platform for compressible electrodes. As the degree of compression increases, the stress of the CF increases substantially. This may be because when the strain is small, the strain mainly comes from the 3D skeleton of the MS; with the strain increases to a larger value (such as $\varepsilon > 70\%$), the stress mainly comes from the compressed CNF/CNT aerogel sheet. The lighting demonstration experiment under compression and release states (Figure 4b and Video S1), and the values recorded using the multimeter (≈5.5 $\Omega$, Figure 4c) further indicate that the CF possesses good structural stability, excellent compressibility, and high conductivity. In contrast, the multimeter value of the MS derivatives (Figure S10) is as high as 14700 $\Omega$, providing sufficient evidence that the CNF/MWCNT carbon aerogel sheet effectively improves the conductivity of the CF platform. Additionally, after being subjected to 10 cycles of 50% compression, the CF retains its 3D continuous porous structure (Figure 4d); furthermore, the aerogel sheets are undamaged (Figure 4e), demonstrating the structural stability of the CF hybrid materials. As shown in Figure 4f, the compressive strain of the PPy—CF and Fe$_3$O$_4$—CF electrodes hardly changes after 10 cycles of 30% compression, with retention rates of up to 99.7%. However, stress intensity losses of 5.0% for PPy—CF and 8.7% for Fe$_3$O$_4$—CF occur. These slight losses are attributed to the multiple soaking steps and the high-temperature and high-pressure treatment (i.e., hydrothermal reaction) during the preparation of the PPy—CF and Fe$_3$O$_4$—CF electrodes, which may have affected the stress intensity. The results described above indicate that the compressible electrodes prepared using the CF as the electroactive material platform are highly durable with reversible compressibility, thus proving its effectiveness as a platform for supporting PPy—CF and Fe$_3$O$_4$—CF electrodes in the assembly of high-performance compressible supercapacitors.

3.3. Electrochemical Evaluation of Electrodes. Considering that compressible supercapacitors require electrodes with excellent capacitive properties, good conductivity, and compressibility, we further evaluated the electrochemical performances of the PPy—CF and Fe$_3$O$_4$—CF electrodes. First, we evaluated the electrochemical performance of the CF platform. As shown in Figure S11a, the cyclic voltammetry (CV) curves exhibit rectangular shapes at a scan rate of 5—100 mV/s, indicating that the PPy—CF electrode possessed good capacitive behavior. All the galvanostatic charge—discharge (GCD) curves (Figure S11b) exhibit classic triangular shapes, revealing the reversible charging/discharging ability of the electrode. Figure 5a,b compares the CV curves of the CF platform, and PPy—CF and Fe$_3$O$_4$—CF electrodes at a scan rate of 100 mV/s, respectively. The current density of the PPy—CF and Fe$_3$O$_4$—CF electrodes is significantly higher than that of the CF platform, which indicates that their capacitance contribution mainly comes from the loaded PPy and nano-Fe$_3$O$_4$. As excepted, the CV curves of the PPy—CF electrode collected at different scan rates exhibit a quasi-rectangular shape (Figure S12a), suggesting that the electrode has ideal capacitive characteristics. Furthermore, all the GCD curves exhibit good symmetry, further confirming the good capacitive behavior of the PPy—CF electrode (Figure S12b). The relationship between the area-specific capacity, capacity retention, and discharge current density of these electrodes is shown in Figure 5c. As expected, the PPy—CF electrode shows high specific capacitance and good capacitance retention. At a current density of 1 mA/cm$^2$, the specific capacitance reached 600.5 mF/cm$^2$ (≈610.1 F/g); at a current density of 30 mA/cm$^2$, the specific capacitance remained relatively high at 406.4 mF/cm$^2$ (≈412.7 F/g). These results correspond to a capacitance retention rate of 67.7%, as shown in Figures 5c and S12c.

To further demonstrate the versatility of the CF platform in the construction of high-performance compressible electrode materials, we prepared the Fe$_3$O$_4$—CF electrode using a similar strategy to match the PPy—CF electrode for assembling a compressible ASC, as noted in Section 2.4 (Supporting Information). The detailed electrochemical characteristics of the Fe$_3$O$_4$—CF electrode are shown in Figure S13a—d in Supporting Information. As expected, the electrode displayed good capacitive properties. At a current density of 1 mA/cm$^2$ (≈0.23 A/g), the specific capacitance reached 727.6 mF/cm$^2$ (≈169.2 F/g); at a current density of 20 mA/cm$^2$ (≈9.2 A/g), the specific capacitance reached 348.5 mF/cm$^2$ (≈81 F/g). To further illustrate the advantages of the CF as a platform, we used the MS derivatives as the platform to introduce electroactive Fe$_3$O$_4$ under the same conditions (details in the Experimental Section). As shown in Figure S14a, the current density of the Fe$_3$O$_4$—CF electrode is significantly higher than that of the Fe$_3$O$_4$—MS electrode, indicating its higher electrochemical capacitance. Meanwhile, the GCD curve of the Fe$_3$O$_4$—CF electrode exhibits a longer discharge time compared to that of the Fe$_3$O$_4$—MS electrode (Figure S14b). This further indicates that the Fe$_3$O$_4$—CF electrode exhibits high capacitance performance. The above results prove that the CF is more advantageous electroactive platform than the MS derivatives. This is mainly because the highly conductive, large, and continuous carbon aerogel sheets (Figure 1e) in the CF platform are more beneficial for active material loading and rapid electron transfer than the narrow skeleton structure of the MS (Figure 1c). In addition, the AC impedance spectra (Figures 5d, S11d, S12d, and S13d) indicate low impedance values of the CF and PPy—CF and Fe$_3$O$_4$—CF electrodes. This was attributed to the conductive 3D network formed by the uniformly dispersed MWCNTs in the carbon aerogel sheets, which were derived from CNF/MWCNT sheets (inset of Figure 1e); this structure effectively increased the conductivity of both the electrodes. Furthermore, the PPy—CF electrode has a smaller AC impedance value than the CF, which could be because of the loaded PPy that further improved conductivity of the PPy—CF electrode. These results further demonstrate the advantages of the CF as an effective electroactive material platform in the construction of high-performance compressible electrodes.
3.4. Performance of the Compressible PPy–CF//Fe₃O₄–CF ASC Device. We evaluated the feasibility of fabricating high-performance compressible supercapacitors with the PPy–CF and Fe₃O₄–CF electrodes prepared on the CF platform by assembling an optimized quasi-solid-state PPy–CF//Fe₃O₄–CF ASC device. Figure 6a shows the CV curves of the PPy–CF positive electrode and the Fe₃O₄–CF negative electrode at a scan rate of 50 mV/s. The stability work windows of the PPy–CF and Fe₃O₄–CF electrodes are 0.0–0.8 and −0.8 to 0.0 V, respectively; this ensures that the assembled device can be operated at working voltages up to 1.6 V. Figure S15a shows the CV curves of the PPy–CF//Fe₃O₄–CF ASC device within a voltage window of 0.8–1.6 V at a scan rate of 50 mV/s. All the CV curves exhibit quasi-rectangular shapes, indicating that the device possesses a wide voltage window and typical capacitive properties. In addition, all the GCD curves (Figure S15b) within the operating voltage range of 0.8–1.6 V exhibit good symmetry, further confirming the good capacitive behavior of the PPy–CF//Fe₃O₄–CF ASC device. As shown in Figure 6b, the CV curves of the PPy–CF//Fe₃O₄–CF ASC device at a working voltage of 1.6 V also exhibit quasi-rectangular shapes under different scan rates; this further demonstrates the excellent capacitive properties and reaction reversibility of the device. Furthermore, the GCD curves of the PPy–CF//Fe₃O₄–CF ASC device at different current densities exhibit typical triangular shapes and long discharge times (Figure 6c), indicating that the device is highly capable of reversible charging/discharging and possesses high specific capacitance. Based on the calculation results obtained from the GCD curves, the PPy–CF//Fe₃O₄–CF ASC device has an excellent mass-specific capacitance and good capacitance retention (Figure 6d). For example, at a current density of 0.2 A/g, a mass-specific capacitance of 172.5 F/g was achieved; furthermore, when the current density was increased to 8 A/g, a relatively high mass-specific capacitance of 103.6 F/g could still be achieved. Figure S15c shows the variations in the mass-specific capacitance and energy density with the working voltage at a current density of 1 A/g. As the working voltage increased from 0.8 to 1.6 V, the mass-specific capacitance and energy density increased correspondingly, with the energy density showing a considerable increase of 54.78%. Figure 6e compares the energy and power densities of the PPy–CF//Fe₃O₄–CF ASC with those of previously reported high-performance compressible supercapacitors. 

Notably, the PPy–CF//Fe₃O₄–CF ASC has outstanding energy and power density values of 59.9 W h/kg (≈ 0.088 MW h/cm²) and 598.4 W/kg, respectively, at a current density of 0.2 A/g. When the current density was increased to 8.0 A/g, the energy and power density values reached 10.1 W h/kg (≈ 0.0156 MW h/cm²) and 12.7 kW/kg, respectively. The energy density values exhibited by the PPy–CF//Fe₃O₄–CF ASC are significantly higher than those exhibited by other high-performance compressible supercapacitors (Table S2), including PANI-SWCNTs–S//PANI-SWCNTs–S (1.28 A/g, 8.8 W h/kg),16 NCF-SSC (1 mA/cm², 1.35 W h/kg),17 F-GRF//F-GRF (2 mV/s, 12.5 W h/kg),19 CNTs/PEDOT sponge–SSC (0.5 A/g, 12.6 W h/kg),35 PPy@CA//PPy@CA (0.5 A/g, 23.8 W h/kg),35 CNT@PPy@MnO₂ sponge//CNT@PPy@MnO₂ sponge (2 mV/s, 8.6 W h/kg),36 δ-MnO₂@CNTs@spponge SSC (0.75 A/g, 28.5 W h/kg),37 NCF//NCG (0.25 A/g, 31.25 W h/kg),38 GASC (1 A/g, 7.99 W h/kg),39 PANI/CC–SSC (0.25 A/g, 16.1 W h/kg),40 3D-CNF//3D-CNF (0.1 A/g, 10.8 W h/kg),41 and PAGH–EC (1 A/g, 26.5 W h/kg).40 As shown in Figure 6f, the PPy–CF//Fe₃O₄–CF ASC retains 95.6% of its initial capacitance after 4000 charge/discharge cycles under different compression conditions, indicating high cycle stability. Moreover, the Coulombic efficiency of the device ranges up to 96.8%. This good cycle performance and
reversible charge–discharge behavior may be ascribed to good structural stability, excellent compressibility, and high conductivity of the CF platform, which can prevent the electrode damage that results from collapse of the structure and the peeling of the electroactive materials during the deformation process.

To show that the PPy–CF//Fe₃O₄–CF-ASC device can satisfy the energy storage needs of portable electronic devices under compressive deformation, we investigated the electrochemical performance of the device under different levels of compression. As shown in Figure 7a, the CV curves under 0–50% compression show similarly sized quasi-rectangular shapes, indicating that the device possesses good capacitive properties and shows stable electrochemical performance under compressive deformation levels of up to 50%. The GCD curves obtained under different levels of compressive deformation offer further evidence of the stable electrochemical performance of the device (Figure 7b). The capacitance retention rates of the device under different compression rates also indicate its excellent stability and capacitive properties (Figure 7c; detailed calculations are shown in Supporting Information). It is evident from the CV and GCD curves that the device achieves capacitance retention rates nearly 100% under different levels of compression.

Additionally, the functional relationship curves of the volumetric specific capacitance versus compressive depression, obtained through experiments and theoretical calculations, roughly coincide with each other (Figure S16); this further indicates that the device could provide stable electrochemical performance under compressive deformation. The Nyquist plot (Figure 7d) shows that minor changes occur with increases in compressive deformation, leading to slight decreases in the charge-transfer resistance ($R_s$) in the high-frequency region. This may be attributed to increased contact within the 3D network-like structure of the CF as the compression level increased, which would cause the formation of more conduction pathways. Notably, the mass-energy density of the device remains unchanged under different levels of compressive deformation (retention rates > 98%), and the measured volume-energy density values under various compressive deformation levels are roughly consistent with the corresponding values obtained from theoretical calculations (Figure 7e); this provides sufficient evidence of the stable energy storage characteristics of the device when subjected to compressive deformation. Furthermore, owing to the particularity of compressible supercapacitors, the electrodes usually have a certain thickness, which results in a small volumetric energy density value of the assembled device. Nevertheless, the
PPy-CF//Fe3O4-CF-ASC device still has high capacity and extraordinary volumetric energy density values. When the compression level is 50%, the volume-specific capacity and energy density values are as high as 443.1 mF/cm³ and 0.139 mWh/cm³ respectively.

To further demonstrate the potential uses of the fabricated device, two compressible PPy-CF//Fe3O4-CF-ASCs with dimensions of 1.5 cm × 1.0 cm × 1.0 cm were connected in series to form an integrated unit for driving electronic devices. As shown in Figure 7f, the integrated unit can light a 3.0 V LED light bulb, indicating that the energy storage device possesses a high energy density and good integrability. The light bulb is powered when the integrated unit is subjected to 40% compression (Figure 7g and Video S2); this shows the stable energy storage properties of the PPy-CF//Fe3O4-CF-ASC even under compressive deformation. In addition, Figure 7h shows that the CV curves of the integrated unit exhibit a working potential range of 3.2 V at a scan rate of 50 mV/s, which is twice the range of a single supercapacitor. The charge/discharge curves of the integrated unit also indicate a maximum charging potential of 3.2 V (Figure 7i). These results demonstrate that the compressible PPy-CF//Fe3O4-CF-ASC devices possess significant potential for applications as energy storage units in portable and wearable electronic devices that are prone to compressive deformation in their working environments.

4. CONCLUSIONS
This study demonstrates a simple and scalable strategy for the preparation of an efficient and versatile CF hybrid material platform through the combination of the CNF/MWCNT aerogel and MS. This platform was intended for preparing electrodes to construct a high-performance compressible supercapacitor. Owing to the 3D network-like structure derived from the carbonization of the MS, which provided a supporting skeleton, the aerogel sheets derived from the carbonization of the CNF/MWCNT sheets had high conductivities and high specific surface areas. Therefore, the CF possessed good compressibility, good structural stability, high conductivity, and a large interfacial contact area; therefore, the CF was a convenient and effective platform for the subsequent introduction of electroactive materials. As expected, the PPy-CF and Fe3O4-CF electrodes prepared using the CF as a carrier showed good capacitive properties and compressibility, with specific capacitances of 600.5 mF/cm² (≈610.1 F/g) and 727.6 mF/cm² (≈169.2 F/g), respectively. In addition, the strain retention rates of both electrodes reached up to 99.7% after multiple compression cycles (ε = 30%). The obtained PPy-CF//Fe3O4-CF-ASC device exhibited reversible charging/discharging at a voltage of 1.6 V as well as specific capacitance and energy density values of up to 172.5 F/g and 59.9 W h/kg, respectively. Good capacitive behavior was maintained along with a capacitance retention of up to 98.3 under 50% compressive deformation. In summary, these results indicate that the assembled device possesses significant potential for applications in compressible, portable, and wearable electronic devices.

ASSOCIATED CONTENT
Supporting Information
The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.1c00952.

Calculation equation, SEM image of the CF with low magnification, CAs; compressive stress—strain curves of the CAs and CF at strains of 30%; adsorption—desorption isotherms and the corresponding PSD curves of MS derivatives and CFs; optical images showing the shape and size changes of MS, MS-derivative CNF/MWCNT/MS hybrid aerogel material, and CF platform; optical images showing the lightweight CF with a size of 2 cm × 2 cm × 1 cm standing on the tip of Dog tail grass; optical image of CF, PPy-CF, and Fe3O4-CF; optical image showing the Fe3O4-CF electrode adsorbed with the magnet under suspended state; full XPS spectra of PPy-CF and Fe3O4-CF electrodes; photograph of measuring bulk resistance of the CF platform with a multimeter; CV curves of the CF electrode, compressible PPy-CF electrode, compressible Fe3O4-CF electrode, and PPy-CF//Fe3O4-CF-ASC device; GCD curves of the CF electrode, PPy-CF electrode, compressible Fe3O4-CF, and PPy-CF//Fe3O4-CF-ASC device; specific capacitance and capacitance retention rate of the CF electrode, PPy-CF electrode, Fe3O4-CF electrode, and Fe3O4-MS electrodes at different current density; specific capacitance and energy density of the PPy-CF//Fe3O4-CF-ASC device under different voltage windows; Nyquist plots of the CF electrode, PPy-CF electrode, Fe3O4-CF electrode, Fe3O4-MS electrode, and PPy-CF//Fe3O4-CF-ASC device; and volume capacitance and volume-energy densities of PPy-CF//Fe3O4-CF-ASC device as a function of strain (PDF)
Demonstration experiment of using the CF platform as a connecting wire to light up an LED bulb under multiple compression conditions (MP4)
LED indicator (3 V) lit by two units of the compressible PPy-CF//Fe3O4-CF-ASC device (size: 1.5 cm × 1.0 cm × 1.0 cm) in series under 40% compressive deformation (MP4)

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Notes

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