**Bifunctional Devices** 



# Rational Design of a Flexible CNTs@PDMS Film Patterned by Bio-Inspired Templates as a Strain Sensor and Supercapacitor

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Flexible devices integrated with sensing and energy storage functions are highly desirable due to their potential application in wearable electronics and human motion detection. Here, a flexible film is designed in a facile and low-cost leaf templating process, comprising wrinkled carbon nanotubes (CNTs) as the conductive layer and patterned polydimethylsiloxane (PDMS) with bio-inspired microstructure as a soft substrate. Assembled from wrinkled CNTs on patterned PDMS film, a strain sensor is realized to possess sensitive resistance response against various deformations, producing a resistance response of 0.34%, 0.14%, and 9.1% under bending, pressing, and 20% strain, respectively. Besides, the strain sensor can reach a resistance response of 3.01 when stretched to 44%. Furthermore, through the electro-deposition of polyaniline, the CNTs film is developed into a supercapacitor, which exhibits a specific capacitance of 176 F  $g^{-1}$  at 1 A  $g^{-1}$  and a capacitance retention of 88% after 10 000 cycles. In addition, the fabricated supercapacitor shows super flexibility, delivering a capacitance retention of 98% after 180° bending for 100 cycles, 95% after 45° twisting for 100 cycles, and 98% after 100% stretching for 400 cycles. The superior capacitance stability demonstrates that the design of wrinkled CNTs-based electrodes fixed by microstructures is beneficial to the excellent electrochemical performance.

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With the rapid development of flexible electronic devices including wearable displays and intelligent bracelets,<sup>[1-3]</sup> flexible energy storage devices including supercapacitors<sup>[4-6]</sup> and batteries,<sup>[7,8]</sup> have attracted intensive interest. Compared with traditional energy storage devices that are bulky and rigid, flexible supercapacitors and batteries usually maintain outstanding electrochemical properties under diverse deformations such as bending, twisting, and stretching. For practical applications, flexible energy storage devices usually require a rational design to be integrated with more functions, e.g., sensing, energy harvesting, or energy transit system.<sup>[9-12]</sup> A self-powered integrated device was fabricated by connecting an asymmetric microsupercapacitor to a TiO<sub>2</sub> photodetector,<sup>[5]</sup> or a self-powered smart cloth was designed by weaving a fiber-based generator into a wireless temperature sensor.<sup>[13]</sup> However, the integrated devices in series face the challenges of extra weight and element size.<sup>[14]</sup>

Strain sensors, widely applied for human motion detection<sup>[15–17]</sup> and health

monitoring systems,<sup>[18-20]</sup> can be operated by recording the change of electrical characteristics, such as the resistance change caused by mechanical deformations.<sup>[21]</sup> Traditional strain sensors based on metals and semiconductors, however, can detect only a very narrow range of strain ( $\varepsilon < 5\%$ ) due to their rigid nature.<sup>[22,23]</sup> Strain sensors with high performances (e.g., high stretchability, high sensitivity, and broad sensing range) can be obtained by employing nanomaterials as conducting components to design the strain-sensing materials.<sup>[24-26]</sup> Among them, carbon nanotubes (CNTs)-based strain sensors have been extensively studied due to their excellent electrical and mechanical properties. They can be assembled in wrinkled structure by prestretching method to realize a highly stretchable strain sensor.<sup>[27–29]</sup> Another efficient strategy to achieve the goal is to introduce the nano-/microstructures (including pyramid arrays,<sup>[30]</sup> microdome arrays,<sup>[31]</sup> microgrooves,[32] etc.) on flexible polymeric substrates via rational nanotechnologies. To date, most of these microstructures were fabricated through traditional lithography technique involving a complicated, time-consuming, and high-cost process.<sup>[33]</sup> To

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address this issue, bio-based materials have been applied as templates to create microstructures in a low-cost and facile fabrication method.<sup>[34–38]</sup> For instance, a flexible polydimethylsiloxane (PDMS) film can be fabricated and tailored with uniform crisscross pattern microstructures from a textile silk template,<sup>[36]</sup> and bionic hierarchical structures from natural leaf template,<sup>[34]</sup> etc. However, researches on bifunctional or multifunctional devices that can simultaneously detect the strain or pressure changes and store energy by a sole active material were rarely reported.

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Herein, a simple and low-cost bio-inspired templating strategy was used to fabricate a flexible bifunctional device. A flexible patterned PDMS film was molded from leaf, followed by paving wrinkled aligned CNTs, which were applied as conductive layer due to their excellent conductive and mechanical properties. The wrinkled CNTs on patterned PDMS film functioned as a strain sensor, which showed sensitive resistance response to various deformations such as bending, pressing, and stretching. The resistance response was also investigated in detecting a series of finger motions. Furthermore, the CNTs-PDMS film was developed into a flexible supercapacitor by depositing polyaniline (PANI), performing a large specific capacitance, superior rate performance, and cycle stability.

The fabrication process of the strain sensor is schematically illustrated in **Figure 1a**. Different from the traditional photolithography strategy involving complex and time-consuming procedure, the bio-inspired architectures here were created simply

by camphor leaves, which are cost-efficient and easy-getting. The microstructure of the leaf can be seen in Figure S1 in the Supporting Information. First, the PDMS precursor was coated onto a leaf which was stuck on the glass slide, and then exposed to air at room temperature to cure the precursor. After removing PDMS film from the leaf, its surface was etched with various textured microstructures by the leaf (Figure 1b and Figure S2). The obtained PDMS film had a thickness of around 900 µm whereas the depth of patterned microstructures by the leaf template was in a range of only several micrometers (<10 µm) as observed from scanning electron microscopy (SEM) (Figure S3, Supporting Information). Therefore, the effect of leaf template is independent with the PDMS thickness. Furthermore, the PDMS film was prestretched at a defined amount of strain and fixed on the glass slide, with 2 layers of CNTs sheets paving along the stretching direction. The prestrain was defined as  $\varepsilon_{\rm pre} = (L_1 - L_0)/L_0$ , where  $L_0$  and  $L_1$  correspond to the length of PDMS before and after stretching. In this work, all the samples were prestrained to 25% as a demonstration. Then, several drops of ethanol were added to ensure the aligned CNTs film adhered firmly on the prestretched PDMS film and can be fixed by the microstructures. Subsequently, the prestretched PDMS film was released with the formation of the wrinkled CNTs film on the PDMS substrate. Figure 1c-e shows SEM images of CNTs sheets with 25% prestrain, demonstrating that a wrinkled structure can be distinguished distinctly. To compare, flat CNTs film was obtained without prestretching process as observed in Figure S4 in the Supporting Information.



Figure 1. a) Schematic illustration of the fabrication of wrinkled CNTs@P-PDMS film. b) A SEM image of P-PDMS film. c-e) SEM images of wrinkled CNT sheets on P-PDMS film in different magnifications.

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**Figure 2.** a) Plots of relative resistance of CNTs@P-PDMS and CNTs@PDMS as a function of tensile strain. b) Photograph of the CNTs@P-PDMS film under stretching. Relative resistance response of CNTs@P-PDMS to 20% strain c), 10 gram weight of pressing d) and bending e), respectively. f) Response signals of CNTs@P-PDMS detecting the finger bending.

The strain sensor derived from CNTs paved on patterned polydimethylsiloxane (P-PDMS) material through a 25% prestretching process was assembled and denoted as CNTs@P-PDMS (Figure S5, Supporting Information). The strain sensing performance was tested according to the relative resistance variation under stretching. The sensitivity of stain sensor is represented by the gauge factor (GF), the slope of the curve in **Figure 2**a, which is defined as  $(\Delta R/R_0)/\varepsilon$ , where  $\Delta R$  refers to the change of the resistance,  $R_0$  refers to the original resistance, and  $\varepsilon$  is the applied strain when the sensor is stretched. In Figure 2a, the relative change of the resistance-strain curve of the CNTs@P-PDMS was divided into three regions with different slopes, showing a GF value of 0.4 (at 0-20% strain), 5.5 (20-35% strain), and 22.6 (35-44% strain). Sensing performance about strain sensors constructed by conductive carbon nanomaterials with PDMS is detailed in Table S1 in the Supporting Information. The above strain sensor (wrinkled CNTs@P-PDMS) still worked at strain of 44%, demonstrating that it possessed a relatively wide sensing range among those strain sensors. In addition, GF values (5.5 and 22.6) at >20% strain illustrates its good sensitivity. The stretching apparatus was shown in Figure 2b. Compared to CNTs-PDMS without leaf template (denoted as CNTs@PDMS) with a resistance response of 1.98 at 44% strain, the CNTs@P-PDMS exhibited a higher value of 3.01 at 44% strain, attributed to the fixing function of the patterned microstructure, thus reducing connection between CNTs. Within the strain of 20%,  $\Delta R/R_0$ increased slightly in a similar linear shape. Over the 20% strain, CNTs@P-PDMS film dramatically increased, due to a reduction in the overlapping area between adjacent wrinkled CNTs. The resistance increased continuously at strains >40%, probably because of the disconnections formed between CNTs during the stretching over the prestrain. To demonstrate the resistance response stability and repeatability of the device, wrinkled CNTs@P-PDMS film was measured under a stepwise tensile strain with 20% stretching and produced a resistance response of 9.1% in Figure 2c. The rapid ascending and descending of the resistance and a stable step demonstrated the high sensitivity and stability. To compare, flat CNTs@P-PDMS film without prestretching was also investigated, showing a narrower sensing range of 25% (Figure S6, Supporting Information), evidencing that the prestretching process endowed the wrinkled CNTs with a higher stretchability, thus widening the sensing range. The resistance response performance of strain sensor with 50% prestrain CNTs-PDMS was also obtained (Figure S7, Supporting Information). Compared with P-PDMS with 25% prestrain, the resistance response plot of P-PDMS with 50% prestrain raised more gently in accordance with the function of prestrain to stabilize the electrical conductivity. Therefore, the PDMS-CNTs film with wrinkled structures and prestrain not only possessed an obvious resistance response to strain change but also allowed the sensor to deliver a wider detective strain range (over 40%) under stretching. However, the traditional strain sensors made from metals or semiconductors can only exhibit narrow sensing ranges (<5%).<sup>[22,23]</sup> This phenomenon proved that the rational design of wrinkled structure fixed by microstructures endowed the film sensor with sensitive electrical conductivity, excellent mechanical stability, and durability under stretching. In addition to the stretching deformation, the strain sensor also responds to the resistance under pressing (0.14%) and bending (0.34%), indicating a versatile application. With a cyclic lifting







**Figure 3.** a) Schematic illustration of the fabrication process of the flexible electrode with the electrodeposition of PANI. b) A SEM image of CNT sheets with 50% PANI. The weight percentage of PANI accounts for 50% of total weight including PANI and CNT sheets. Electrochemical performance of the PANI-CNTs@P-PDMS supercapacitor with 50% PANI: c) CV curves at a scan rate of 10 to 100 mV s<sup>-1</sup>, d) charge–discharge curves at current densities from 0.5 to 5 A g<sup>-1</sup>, e) cycling measurement at 1 A g<sup>-1</sup>. f) Charge–discharge curves of supercapacitors connected in parallel at 50 mV s<sup>-1</sup>. g) CV curves of supercapacitors connected in series at 1 A g<sup>-1</sup>. The insets are circuit diagrams. h) Photographs of four supercapacitors connected in series to achieve a high potential to illuminate a red LED.

and letting down of a 10 gram weight to press and release the CNTs conductive layer, the sensor demonstrated an instant resistance change of 0.14% (Figure 2d). Besides, owing to the excellent flexibility and mechanical robustness of the PDMS substrate, the reversible cyclic bending/unbending can be measured, showing a resistance response of 0.34% (Figure 2e). To illustrate the potential application of motion detection and health monitoring, the strain sensor was attached to the finger. During the finger-bending process, the sensor was deformed with different bending angles, leading to a change of the relative resistance response as shown in Figure 2f. Besides, the thickness of CNTs can be tuned by controlling the stacked layer number of CNTs sheets. The electrical conductivity of CNTs with different layers of sheets has been characterized, exhibiting an average resistance of 9, 4, and 0.8 k $\Omega$  referring to 1, 2, and 10 layers of CNTs sheets in a size of 2 cm (aligned direction)  $\times$  0.5 cm. The resistance response of CNTs in two or ten layers produced a wider sensing range (over 40% strain) than that of one layer (about 32% strain) (Figure S8, Supporting Information), which can be explained by the alleviated deformation of CNTs in stacked multilayers under stretching.

As mentioned above, the CNTs@P-PDMS films exhibit superior electrical and mechanical properties, which can be further developed into flexible and stretchable supercapacitors. Here, PANI was applied as a pseudocapacitive active material due to its outstanding theoretical capacitance and low cost.<sup>[39,40]</sup> As shown in Figure 3a, on the basis of the above CNTs@P-PDMS film, PANI was electrodeposited onto CNTs sheets. The mass of the deposited PANI can be controlled by the deposition time. After introduction of PANI, PANI-CNTs film still had the resistance response under stretching, showing a resistance response of 2.09 under 29.0% strain (Figure S9, Supporting Information). Then PVA-H<sub>3</sub>PO<sub>4</sub> gel electrolyte was uniformly coated on the PANI-CNTs film. Subsequently, the other PANI-CNTs thin film was paralleled to assemble all-solid-state supercapacitor with sandwiched structure. Typical SEM images of CNTs sheets deposited with 50% PANI were presented in Figure 3b and Figure S10 in the Supporting Information, showing a uniform layer deposited all over CNTs sheets without the aggregation of PANI, allowing for the supercapacitor with stable electrochemical performance. The deposited PANI layer was in a thickness of several nanometer as observed from transmission electron

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microscope (TEM) (Figure S11, Supporting Information). In addition, SEM images of 20%, 70%, and 85% PANI were also provided (Figure S12, Supporting Information). To confirm the existence of CNTs and PANI, Raman spectra were characterized (Figure S13, Supporting Information). For bare CNTs, the peaks of 1360 and 1590 cm<sup>-1</sup> were ascribed to D band and G band, respectively.<sup>[41,42]</sup> The peak intensity of G band was much higher than the D band, indicating a good electrical conductivity. The peak at 1200 cm<sup>-1</sup> was assigned to C–H bending of the quinoid ring, 1263 cm<sup>-1</sup> was ascribed to the C–H bending of the benzenoid ring, and 1338 cm<sup>-1</sup> was attributed to C–N+ stretching. The 1490 and 1631 cm<sup>-1</sup> characteristic peaks were attributed to C = C stretching of the quinoid ring and C–C stretching of the benzenoid ring.

To evaluate the electrochemical properties of the assembled supercapacitors, cyclic voltammetry (CV), galvanostatic chargedischarge, and cycling measurements were tested. As shown in Figure 3c, CV curves of the supercapacitor with 50% PANI exhibited a stable quasi-rectangular shape at a scan rate from 10 to 100 mV s<sup>-1</sup> and no obvious deviation appeared even at a high scan rate (e.g., 100 mV s<sup>-1</sup>), indicating a stable rate performance. As a comparison, CV curves of bare CNTs and 20%, 70%, and 85% PANI were also provided (Figures S14 and S15, Supporting Information). The bare CNTs exhibited a typical rectangular shape, indicating an ideal double layer capacitor behavior. The distortion of the rectangular shape and the current density continuously enlarged from 20% to 85% PANI, demonstrating the pseudo-capacitance property originating from PANI material. No obvious redox peaks appeared owing to the relatively low ionic conductivity of the PVA-H<sub>3</sub>PO<sub>4</sub> gel electrolyte. The charge-discharge curves of the supercapacitor were in a nearly symmetrical triangle at the current density from 0.5 to 5 A g<sup>-1</sup>, illustrating the supercapacitor can adapt to broad current densities and retain high reversibility during the charge-discharge process. In addition, the long discharge time also evidenced that the introduction of 50% PANI provided a high specific capacitance (176 F  $g^{-1}$  at 1 A  $g^{-1}$ , or 14.7 mF  $g^{-1}$ and 13.9 mF cm<sup>-3</sup> calculated from the total mass and volume of electrodes) (Figure 3d). The rate performance of the device with different weight percentages PANI was shown in Figure S16 in the Supporting Information. By increasing the current density from 0.5 to 5 A  $g^{-1}$ , the supercapacitors with 0%, 20%, and 50% PANI maintained a stable capacitance. Nevertheless, 70% PANI supercapacitor began to decay from 243 F g<sup>-1</sup> at 0.5 A  $g^{-1}$  to 220 F  $g^{-1}$  at 5 A  $g^{-1}$  and decay trend became even more pronounced in 85% PANI supercapacitor (from 330 F g<sup>-1</sup> at 0.5 A  $g^{-1}$  to 274 F  $g^{-1}$  at 5 A  $g^{-1}$ ). In contrast, although 70% and 85% PANI supercapacitors had advantages in capacitance performance, they were short of stable electrochemical performance. Through the total 5000 cycle measurement process at 1 A g<sup>-1</sup>, the supercapacitors with bare CNTs (Figure S14c, Supporting Information), 20% (Figure S15c, Supporting Information) maintained 100% capacitance retention, demonstrating a stable cycling performance. In addition, the specific capacitance of 50% PANI supercapacitor (Figure 3e) preserved 88% capacitance retention after 10 000 cycles. However, the capacitance retention curves of the supercapacitors with 70% and 85% PANI slipped down dramatically and only 58.2% and 51.7% capacitance were retained respectively after 5000 cycles

(Figure S15f,i, Supporting Information), which were attributed to the poor cycle stability of the aggregation PANI, in accordance with SEM morphology. The electrochemical performance (e.g., specific capacitance and cycle stability) of flexible supercapacitors using PANI and carbon nanomaterials as active materials in previous literature is also summarized in Table S2 in the Supporting Information. Supercapacitors based on 50%PANI-CNTs in this work showed a comparable result (176 F g<sup>-1</sup> at 1A g<sup>-1</sup>, 88% capacitance retention after 10 000 cycles) among previous literature data, demonstrating that the resultant supercapacitor has a potential application in flexible energy devices.

CNTs sheet itself is well known for its excellent conductivity and electrochemical stability.<sup>[43]</sup> Although the introduction of PANI could provide a high pseudocapacitance due to the faradaic reaction, it usually has drawbacks in cycle stability.<sup>[44]</sup> Thus, a rational weight ratio of PANI and CNTs and structure design of the device play the key roles to achieve a high specific capacitance and stable cyclic performance. Based on the above electrochemical measurements, the supercapacitor with 50% PANI revealed both of the high specific capacitance and cycle stability, thus chosen for further investigation. Several supercapacitors can be connected in parallel or series to enhance the capacitance (Figure 3f) or operating voltage (Figure 3g). The operating voltage of four supercapacitors connected in series can be improved to 3.2 V (Figure 3g), which can light a red light-emitting diode (LED) after being charged (Figure 3h), demonstrating the practical application of the supercapacitors for flexible energy storage devices.

Stable electrochemical performance under severe deformations including bending, twisting, and stretching is a critical parameter for flexible supercapacitors. Detailed photographs of the bending and twisting process were provided in Figure S17 in the Supporting Information. CVs of the supercapacitors in different bending angles exhibited no obvious distinction in terms of the rectangle shape at 50 mV  $s^{-1}$  (Figure 4a). CV stability can be retained even when the supercapacitor was bent at 180° and returned to its original state. To further investigate the electrochemical performance in bending deformation, long-term performance under a bending angle of 180° was also measured. The supercapacitor kept up to 98% capacitance retention after 180° bending for 100 cycles (Figure 4b). In case of twisting, the shape of charge-discharge curves only showed a slight difference, indicating the electrochemical and mechanical stability of the device under twisting from  $0^{\circ}$  to  $90^{\circ}$  and returning to the original state (Figure 4c). Besides, the performance of supercapacitor was well maintained with a capacitance retention of 95% after 45° twisting for 100 cycles (Figure S18, Supporting Information). Last but not least, to prove the key function of the leaf template, a comparison between supercapacitors using P-PDMS and smooth PDMS in stretchability was made. Within the 70% strain, both of these two supercapacitors with wrinkled structure had stable performance under stretching (Figure S19, Supporting Information), indicating the synergetic interaction between the CNTs and PANI. It is worth noting that, when the supercapacitor was stretched to 100%, the function of the leaf template played a role (Figure 4d). During the continuous stretching process, the capacitance retention for the supercapacitor with P-PDMS etched by leaf template maintained 98%







**Figure 4.** Electrochemcial performance of the PANI-CNTs@P-PDMS supercapacitor under bending, twisting, and stretching deformations. a) CV curves of the supercapacitor under different bending angles from 0° to 180° at 50 mV s<sup>-1</sup>. b) Dependence of capacitance retention on bending/ unbending number. c) Charge–discharge curves of the supercapacitor at different twisting angles at 1 A g<sup>-1</sup>. d) Capacitance retention of the supercapacitors (PANI-CNTs@P-PDMS) under 100% strain as a function of cycle number.

after 400 cycles. However, the supercapacitor using smooth PDMS without leaf etching worked in failure early (at the 30th stretching cycle), which demonstrated unstable electrochemical performance. This result further proves the bio-inspired micro-structures produced by leaves are vital to reducing the strain distribution<sup>[45]</sup> and keep the CNTs-based electrodes in constant distance during stretching/releasing process, thus enhancing the stability of supercapacitor performance.

In summary, a bifunctional device was realized via bio-inspired templating method. By leaf template etching and prestretching process, a strain sensor based on wrinkled CNTs suspended on a flexible patterned PDMS substrate was fabricated and demonstrated an obvious resistance response under bending (0.34%), pressing (0.14%), and stretching (9.1%). As a strain sensor, a resistance response of 3.01 was obtained at 44% strain. Furthermore, the CNTs film was developed into a stretchable flexible supercapacitor by introducing electroactive material PANI onto CNTs. The assembled supercapacitor with 50% PANI possessed a specific capacitance of 176 F  $g^{-1}$  and a capacitance retention of 88% after 10 000 cycles. Under severe deformations, the supercapacitor showed stable electrochemical properties, maintaining 98% and 95% capacitance after 100 cycles under 180° bending and 45° twisting, respectively. Especially, the stretchable supercapacitor with PANI-CNTs on patterned PDMS operated superior to that of smooth PDMS during the repeated stretching at 100% strain and delivered a capacitance retention of 98% after 400 cycles, indicating the fixing function of bio-inspired architecture created by leaf template. All in all, our fabricated device

derived from the rational design of flexible materials exhibits excellent sensing and energy storage functions, providing a promising design to realize multifunctional devices.

#### **Experimental Section**

*Materials*: Aniline ( $\geq$ 99.5% AR grade) was purchased from Aladdin. PDMS precursor was purchased from Dow Corning. Poly(vinyl alcohol) (PVA, Mw = 85 000 to 124 000) was purchased from Sigma-Aldrich. Ethanol ( $\geq$ 99.7% AR grade) and H<sub>3</sub>PO<sub>4</sub> ( $\geq$ 85% GR grade) were purchased from Sinopharm Chemical Reagent Beijing Co., Ltd. H<sub>2</sub>SO<sub>4</sub> (95.0–98.0% AR grade) was purchased from Shanghai Ling Feng Chemical Reagent Co., Ltd. All chemicals were used as received. The deionized water was acquired from a Millipore Direct-Q3 system.

Preparation of P-PDMS films: The leaves picked from camphor trees were first washed with deionized water and then dried in air. To prepare the PDMS film patterned by the leaf, a mixture of PDMS precursor and curing agent at a weight ratio of 10:1 was coated onto the leaf, which was attached to the glass slide with a double-faced adhesive. The thickness of PDMS film can be varied by controlling the coating volume of the mixture. Here, 0.7 mL of the mixture was poured onto a glass slide with a size of 3.5 cm  $\times$  2.5 cm. After curing at 70 °C for 2 h in oven, the PDMS film was peeled off from the leaf and obtained with patterned microstructure. Finally, the resulting P-PDMS film was in a thickness of 0.8–0.9 mm and cut into 0.5 cm  $\times$  1.4 cm in size for use.

Fabrication of Strain Sensors Based on CNTs@P-PDMS Films: To fabricate wrinkled CNTs@P-PDMS films, the aligned CNTs sheets were obtained from spinnable CNTs arrays through a dry-drawing process, and paved onto P-PDMS film which was prestretched to 125% of its original length (prestain 25%). CNTs sheets could be further stacked into multilayers



along the same direction. Two layers of CNTs sheets were applied in the work. Here, the synthesis of spinnable CNTs arrays had been reported previously,<sup>[46,47]</sup> and the density of CNTs sheets was 1.41 ug cm<sup>-2</sup>. Then, the prestretched PDMS film was released with the formation of the wrinkled CNTs. Finally, a strain sensor was assembled by using copper wires to connect two ends of the CNTs@P-PDMS film with silver paste.

Fabrication of Supercapacitors Based on PANI-CNTs@P-PDMS Films: The above-fabricated wrinkled CNTs@P-PDMS film can be further developed into a supercapacitor. Electrodeposition of PANI on the CNTs@P-PDMS film was performed in a three-electrode system using a platinum plate and Ag/AgCl (saturated KCl) as the counter and reference electrodes, respectively. Prior to electrochemical deposition process, CNTs sheets on P-PDMS were dipped in the electrolyte for 5 min. The electrodeposition process was conducted at 0.75 V (vs Ag/ AgCl) in 100 mL solution containing 0.1 M aniline and 1.0 M H<sub>2</sub>SO<sub>4</sub>. After electro-polymerization, the composite film was put into a beaker filled with deionized water for several seconds to wash away the impurities. Finally, a symmetric supercapacitor was assembled by coupling two pieces of PANI-CNTs@P-PDMS films coated with PVA-H<sub>3</sub>PO<sub>4</sub> gel electrolyte uniformly. The PVA-H<sub>3</sub>PO<sub>4</sub> electrolyte was prepared by dissolving PVA powder (0.5 g) into deionized water (5 mL) and H<sub>3</sub>PO<sub>4</sub> (0.5 g). The mass of deposited PANI had been calculated from the total Faradic charge during the electro-polymerization process.

Characterization, Sensor, and Electrochemical Measurements: The microstructure of the samples was characterized by using fieldemission scanning electron microscope (FE-SEM) (JEOL, JSM-7800F, operated at 1 kV) and transmission electron microscope (JEOL 2100, operated at 200 kV). Raman spectroscopy was performed using a 488 nm laser (WITEC, Alpha300M+). The response of the sensor to the strain was measured by the tensile tester (HY-0350, Shanghai Heng Yi Precise Instrument Co., Ltd) at a constant loading speed of 10 mm min<sup>-1</sup>. The resistance variation of the sensor was detected at 3 V by the SourceMeter (Keithley 2400) using a two-point probe method and the data were recorded by the LabVIEW program. The electrodeposition, galvanostatic charge/discharge, and cyclic voltammetry performance were conducted on an electrochemical station (Chenhua, CHI 660E) at a voltage window of 0-0.8 V. The long-term charge/discharge measurements were performed on the system (NEWARE, CT-4008) at a current density of 1 A g<sup>-1</sup>. The capacitance of the PANI-CNTs@ PDMS supercapacitor was also measured under a long-term stretching process conducted by the tensile tester at a loading speed of 5 mm min<sup>-1</sup>. The specific capacitance of the device was obtained according to the galvanostatic discharge curves, calculated by the equation as follows: C =  $2it/m\Delta V$ , where *i* and *t* correspond to the discharging current and time, m is the mass of the CNTs and PANI in the electrode, and  $\Delta V$  is the potential window range of the discharging curve.

### **Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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## **Conflict of Interest**

The authors declare no conflict of interest.

## **Keywords**

bio-inspired templates, flexible supercapacitors, microstructures, strain sensors

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