Supplementary Materials for

Fixture-free omnidirectional prestretching fabrication and integration of crumpled in-plane micro-supercapacitors

Ying Wang et al.

Corresponding author: Liangti Qu, lqu@mail.tsinghua.edu.cn; Yang Zhao, yzhao@bit.edu.cn; Feng Liu, liufeng@imech.ac.cn

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Section S1. Materials and Methods

S1.1 Materials

Kapton tape was purchased from Shenzhen Jiesheng Electronic Material Co., Ltd. Carboxylated multiwalled carbon nanotubes was purchased from Aladdin. Carbon nanotube and graphene aqueous inks (solid CNTs/graphene content: 5wt%; additive: PVP (1.25wt%); solvent: deionized water) were purchased from Suzhou Tanfeng Graphene Technology Co., Ltd. Graphite painting was purchased from Shenzhen Jingzhe Economic Co., LTD. Commercial balloons with different size were obtained from the local market. Poly(vinyl alcohol) and sulfuric acid were purchased from Aladdin. Potassium ferricyanide was purchased from InnoChem.

S1.2 Preparation of CNTs ink

First, 10 mL of sodium alginate (5 mg mL⁻¹) or polyvinyl pyrrolidone (PVP, 2 mg mL⁻¹) adhesive aqueous solutions were prepared by stirring the deionized water and sodium alginate (or PVP) mixture at room temperature. Then 100 mg carboxylated CNTs powder was added to the prepared adhesive mixture solution, and uniform CNT paste was obtained by ultrasonic and stirring for 4 hours at room temperature. The concentration of CNTs can be fluctuated based on this preparation scheme to obtain the optimal paste. However, in order to shorten the fabrication period of the device, the similar commercial CNTs ink was chosen for subsequent device design and exploration.

S1.3 Evaluation the interdigital electrode mass

In the case of electrode prepared by commercial CNT ink, the overall CNT ink density was first evaluated by weighing a certain volume of CNT ink, and its density was calculated as 1.1 g mL⁻¹. Since the interdigital groove area of the PI electrode mask is 13.9 mm² and the thickness of the PI mask is 50 μm, and the volume of CNT ink filling the mask groove is exactly the same as that of the mask groove, thus the volume of CNT ink used as the electrode is calculated as 0.695 mm³. According to the overall CNT ink density and solid CNT content (5wt%), the final interdigital electrode mass is roughly estimated to be 1.1 g mL⁻¹ × 0.695 mm³ × 5wt% = 0.75 mg. After complete drying, the final interdigital electrode with thickness of 2 μm, area of 13.9 mm² and mass loading of ~0.75 mg was obtained.

For the devices with stacked structure, the specific mass loading of each device at various stacked electrodes under different simulative compress strains is evaluated to be ~0.75 mg. To eliminate the contribution of crumpled structures and effects of mass loading, three stacked in-plane devices with similar mass loading under different strains (20%, 40%, 80%) were fabricated, and named as stacking-20%, -40%, -80%. Since the area of the device
changes under different strains, the similar mass loading of each device (such as stacking-20%, -40%, -80%) was achieved by adjusting the thickness of microelectrodes. Specifically, we prepared electrode masks of different sizes according to the actual device sizes under different strains (20%, 40% and 80%), and controlled the electrode thickness by alternate scraping and drying process to achieve the same quality of CNT ink into the mask grooves with different sizes. Eventually, the devices of stacking-20%, -40%, -80% with flat electrode structure were obtained.

**S1.4 Preparation of Electrolytes**

PVA/H$_2$SO$_4$ hydrogel electrolyte was obtained by mixing 98% H$_2$SO$_4$, PVA, and deionized (DI) water with a mass ratio of 1:1:10, and then heating at 80°C for 3 h until it became clear. The artificial seawater electrolyte consisted of 26.5 g L$^{-1}$ NaCl, 24 g L$^{-1}$ MgCl$_2$, 0.73 g L$^{-1}$ KCl, 3.3 g L$^{-1}$ MgSO$_4$, 0.2 g L$^{-1}$ NaHCO$_3$, 1.1 g L$^{-1}$ CaCl$_2$ and 0.28 g L$^{-1}$ NaBr. For the MSCs as flight power supply system, 1M MnSO$_4$/ZnSO$_4$ gel-electrolyte was prepared to ensure that the integrated device can work in the air for a long time.

**S1.5 Electrochemical Measurements**

All electrochemical measurements were performed by a CHI760D workstation. For the electrochemical property of the devices with different size, the CV measurements with scan rates from 10 to 200 mV s$^{-1}$ and GCD profiles conducted at current densities ranging from 0.004 to 1 mA cm$^{-2}$ were evaluated with PVA/H$_2$SO$_4$ as electrolyte. Electrochemical impedance spectra (EIS) were recorded in the frequency range from 100 kHz to 0.01 Hz with the AC amplitude of 5 mV. Cycling stability was carried out at a current density of 0.08 mA cm$^{-2}$. The area capacitance was calculated based on the area of the whole device except for the current collector exposed parts. High-voltage MSCs consisting of 108 units in series were tested by Keithley Model 2600A.

**S1.6 Electrochemical activity test**

Electrochemical activity measurements were performed in a three-electrode system with Pt as a counter electrode (CE), and Ag/AgCl electrode as a reference electrode (RE). The CNT films on the balloon surface (with an initial size of 1.5×1.5 cm) were shrunk at 0%, 20%, 40% and 80% strain, respectively, and cut down as the corresponding working electrodes (WE). K$_3$[Fe(CN)$_6$] (12 mM) was dissolved in 0.5 M phosphate buffer as the electrolyte. The electrolyte was degassed for 20 min before each measurement to avoid the currents related to O$_2$ reduction. CV measurements with a scan rate of 50 mV s$^{-1}$ and EIS were recorded in the frequency range from 100 kHz to 0.01 Hz with the AC amplitude of 5 mV.

**S1.7 Mesoscopic transport model**
A tight adsorption model is applied to investigate the conductivity of van der Waals contacted CNTs, and according to Reich model,(42) CNT Hamiltonian is expressed as \( \text{H}=\text{H}_1+\text{H}_2+\text{H}_{\text{int}} \), where \( \text{H}_1 \) and \( \text{H}_2 \) are Hamiltonians for the first and second layer of graphene, and \( \text{H}_{\text{int}}=\gamma r e^{-|\text{d}_{ij}|/\beta} \) represents the interaction between two layers. Here \( r \) vector points from the ist atom to the jst atom, and \( d \) vector with its magnitude equals the interlayer distance and its direction pointing from the graphene layer including ist atom to another graphene layer including jst atom, and \( \gamma, \beta \) are hopping parameter and characteristic distance, respectively. The definition and values of all parameters could be found in Reference 29 This model could be extended for multilayer graphene, i.e. \( \text{H}=\sum_i \text{H}_i+\sum_{ij} \text{H}_{\text{int},ij} \), where \( i,j=1,\ldots,N \) and \( |i-j|=1 \) as demonstrated in Reference 30. The recursion Green’s function method(44) is used to calculate the conductance. The advanced Green’s function of the conductor region is given as

\[
G_C=\lim_{\eta \rightarrow 0^+} \left[ (E+i\eta)I-\Sigma_L^{\dagger}\Sigma_R \right]^{-1}
\]

where \( \Sigma_L \) and \( \Sigma_R \) represents self-energies of left and right leads, and \( \text{H}_C \) is the Hamiltonian of the conductor region. The differential conductance at given energy \( E \) is written as

\[
G(E)=\frac{2e^2}{h} \text{Tr} \left( \Gamma_R G_C \Gamma_L G_C^{\dagger} \right)
\]

where \( \Gamma_{L,R}=i[\Sigma_{L,R}^{\dagger}\Sigma_{L,R}] \) are broadening matrices. More conductance calculation details could be found in our previous paper Reference 30.

Section S2. Supporting Notes

S2.1 Characterization of CNT film

The uniform CNT with hundreds of nanometers in length and about 15 nm of diameter can be seen in fig. S2, A and B. XRD and XPS spectra (fig. S2, C and D) reveal the obvious characteristic peak of C, and the EDS result (fig. S2E) exhibits high levels of carbon element, illustrating the negligible impurities in CNT paste.

S2.2 Determination of the optimal CNT ink concentration for device fabrication.

By using the originally CNT ink (solid CNTs content: 5wt%) as the raw material for interdigital electrodes, the contracted microelecrode reveals short circuit phenomena and poor shape retention due to the excessive electrode material (fig. S3A). We further diluted the original CNT ink by 1.2, 2, 4 and 8 times named D-1.2, D-2, D-4 and D-8, respectively, and obtained the corresponding crumpled interdigital microelectrodes. As shown in the fig. S3, B–E, all of the microelectrodes prepared by the different concentration of CNT inks maintain
good interdigital structure without short circuit phenomenon. Besides, there is no significant difference in the overall size of these devices (fig. S3, B–E), but only the wavelength of the induced folds decreases with the decrease of CNT concentration (fig. S3, F–I). Eventually, the electrochemical test shows that D-1.2 device possessed the highest areal capacitance, thus the CNT ink is determined to be D-1.2.

**S2.3 Tests of adhesion between CNTs and balloons**

Here, the commercial CNT aqueous-ink is chosen as the electrode material for the following two reasons. (1) Due to the outstanding conductivity, CNTs are the suitable electrode materials for the energy storage system without current collector. (2) CNT with 1D microstructure can maintain good mechanical and electrical properties under external stress, and occur the corresponding reversible deformation. Notably, in order to make the CNT ink evenly and firmly contact with the surface of the balloon, the surface of the inflated balloon was first rubbed with a piece of paper before the CNT ink was coated. After friction, the inflated balloon brings abundant charges on its surface, while the CNT aqueous-ink will carry the opposite positive charges as it approaches the balloon surface. Then, the generated interface electric-field would enhance the binding force and distribution uniformity of aqueous CNT ink (fig. S4A), and the water droplet on the friction-treated balloon surface reveals a larger contact area (fig. S4B) than the one without friction treatment (fig. S4C). To further verify this conjecture, the friction-treated balloon was placed close to the water flow and the electroscope, both the water flow and the electroscope pointer were shifted, indicating that they carried opposite charges (fig. S4, D and E and Movie S1).

However, although the electrostatic force is beneficial to the stable adhesion between the carbon nanotubes film and the balloon during and after shrinkage, the PVP adhesive in the CNT ink plays a dominant role. The adhesion between CNT film and balloon was tested in plane, inflatable balloon and shrunken balloon. On the loose plane of the balloon, the significant peeling CNT materials can be seen on the tape (fig. S4F), while a small amount of shedding CNT materials can be seen in the condition of frictional inflatable balloon (fig. S4G), illustrating the electrostatic action makes the CNT bond to the balloon stronger. After shrinkage, negligible peeling CNT material can be seen on the tape after one debonding, even only a small amount of shedding CNT material after 100 times of debonding (fig. S4H). As shown in Fig. S4I, the inside surface of the balloon with a crumpled interdigital electrode on the outer surface shows an apparent indentation with interdigital shape, which is caused by the remarkable adhesive force between CNT film and balloon. Ultimately, the adhesion strength between CNT film and balloon at 0% and 80% contraction strain are measured to be 0.032
and 0.328 Mpa, and the significantly increased adhesion strength under high strain (80%) further proves the strong adhesion between the balloon and CNT film.

**S2.4 Inference of wrinkling behavior on CNT film by Neo-Hookean bilayer model**

The model of CNT film on the balloon surface under 0% strain is shown in fig. S7A, the thickness of CNT film is much smaller than that of the balloon. After the compression stress is applied, more than 40% compressive strain could lead to computation non-convergence problem for 3D Neo-Hookean bilayer model (fig. S7B). Next, the 2D Neo-Hookean bilayer model will be used to study wrinkling behavior under large deformation. Note that 2D and 3D Neo-Hookean bilayer models share the same physical origin for wrinkling, it is thus believed 2D Neo-Hookean bilayer model could qualitatively or even semi quantitatively describe the wrinkling behavior.

**S2.5 FEM calculation**

In fig. S9, the FEM obtained wavelength variation with respect to compressive strain is compared with the experimental measurement. It could be seen that the onset of wrinkling in FEM (around 35% strain) is much earlier than 20% in the experiment, and on the other hand the onset wrinkling wavelength is quite different. This inconsistency mainly stems from dimensionality, to be specific, in 3D Neo-Hookean bilayer model the wrinkling occurs at 26% strain (closer to experimental observation) and the corresponding onset wrinkling wavelength is about 25 μm, which is comparable to the 10 μm wavelength measured in experiments. The left inconsistency is very limited suggesting the simple Neo-Hookean bilayer model is good enough to reproduce the wrinkling phenomenon.

**S2.6 Calculation of IR_{drop} of the MSC**

The $IR_{drop}$ is calculated from the following equations:

$$IR_{drop} = a + bI$$  \hspace{1cm} (S3)

where $a$ is the difference between the applied and real voltage of MSC; $b$ is the twice of the equivalent series resistance (ESR) of MSC.

**S2.7 Ion diffusion coefficient**

For the typical porous CNT-based electrode, its equivalent series resistance ($R_{ESR}$) is composed of three parts: intrinsic ohmic resistance ($R_\Omega$), interfacial charge transfer resistance ($R_{ct}$), and Warburg diffusion resistance ($R_w$). Among them, $R_w$ comes from the resistance of ion diffusion, and the ion diffusion coefficient can be calculated by using the following equation:
\[ D = \frac{R^2 T^2}{2n^4 F^4 A^2 C^2 \sigma^2} \]  

where \( D \) (cm\(^2\) s\(^{-1}\)) is the ion diffusion coefficient, \( R \) (J mol\(^{-1}\) K\(^{-1}\)) is the gas constant, \( T \) (K) is the absolute temperature, \( n \) is the electron number involved in the redox reaction, \( A \) (cm\(^2\)) is the geometric area of electrode, \( F \) (C mol\(^{-1}\)) is the Faraday constant, and \( C \) (mol cm\(^{-3}\)) is the molar concentration of ions. Additionally, \( \sigma \) (\( \Omega \) s\(^{-1/2}\)) is the Warburg coefficient, which can be calculated by plotting \( Z' \) against \( \omega^{-1/2} \) on the basis of the equation of \( Z' = R_\Omega + R_{ct} \sigma \omega^{-1/2} \). The \( \sigma \) value of 80% is calculated to be 22.4, much lower than that of 40% (47.8), 20% (59.6) and 0% (78.6). Thus, the D value of 80% strain is almost 3.5 times higher than that of 0% strain, which is beneficial for the rapid diffusion for electrolyte ions, ascribing prominent rate capability (fig. S14).

**S2.8 Energy/power densities and cycling stability**

The calculations of areal capacitance, energy density, and power density are followed by our previous papers, and the areal capacitance \( C_a \) was calculated as follows.

\[ C_a = \frac{I(A) \times \Delta t(s)}{\Delta V(V) \times A(cm^2)} \]  

where \( C_a \), \( I \), \( \Delta t \), \( \Delta V \), and \( A \) are the areal capacitance (F cm\(^{-2}\)), charge/discharge current (A), discharge time (s), discharge voltage (V), and the area of the electrode (cm\(^2\)), respectively.

**Calculation of energy density and power density**

\[ E = \frac{I(A) \times \int_{t_b}^{t_e} V(t) \times dt}{A(cm^2) \times 3600} \]  

\[ P = \frac{E(Wh/cm^2)}{\Delta t(s)} \times 3600 \]

where \( E \) and \( P \) are the energy density (Wh cm\(^{-2}\)) and power density (W cm\(^{-2}\)), respectively. \( t_b \) and \( t_e \) are the beginning and ending times of discharging; \( V(t) \) is the device voltage; \( \int_{t_b}^{t_e} V(t) \times dt \) is the integrated area from the galvanostatic discharging curves.

**S2.9 Deduction for the contribution rate of electrochemical performance improvement**

For the stacked electrode structures, the device capacitances of stacking−20%, stacking−40% and stacking−80% are calculated as 3.20, 3.90 and 5.26 F cm\(^{-3}\), which have the improvement of 0.61, 1.31 and 2.67 mF cm\(^{-3}\) in comparison with the initial device (at 0% strain, 2.59 F cm\(^{-3}\)). And the appropriate capacitance enhancement is mainly due to the size minification of devices rather than crumpled structures. For the strain-induced crumpled
devices, both the device size change and generated crumpled structure are the two dominant factors for performance improvement. The capacitances of the MSCs at 20%, 40% and 80% strain are calculated as 3.64, 5.14 and 9.30 F cm\(^{-3}\), respectively, which have the improvement of 1.05, 2.55 and 6.71 mF cm\(^{-3}\) in comparison with the initial device. Therefore, the difference between the enhanced capacitance of the strain-induced devices and the stacked devices can be regarded as the performance improvement caused by the crumpled structures, which are 0.44, 1.24 and 4.04 mF cm\(^{-3}\). Eventually, the contribution ratio of device size change and derived crumpled structures for enhanced capacitance are calculated as 58%, 51%, 40% and 42%, 49%, 60% for MSCs at 20%, 40%, 80% strain (Fig. 3B).

**S2.10 Effective active surface areas (EASA)**

The average \(C_{dl}\) of CNT films at 0%, 20%, 40% and 80% are 0.5, 3, 5 and 11 mF cm\(^{-2}\) respectively. The EASA can be calculated as:

\[
\text{EASA} = \frac{C_{dl}}{C_s}
\]

\(C_s\) is the specific capacitance value for a flat standard with 1 cm\(^2\) of real surface area. The general value for is between 20 µF cm\(^{-2}\) and 60 µF cm\(^{-2}\). Here we use 40 µF cm\(^{-2}\) as the average value. Thus, the EASA for strain of 0%, 20%, 40% and 80% can be obtained as 12.5, 75, 125 and 275 cm\(^2\) (fig. S23).

**S2.11 MD simulation**

The initial CNT film configuration is shown in fig. S18, which is randomly generated and followed by a relaxation and almost all CNTs keep straight. Biaxial compression is applied to simulate the quasi-plane strain state of wrinkling. With compression increasing, CNTs pack more closely, and bending deformation of individual CNT becomes more severely.

**S2.12 Mesoscopic transport theory.**

Here, mesoscopic transport theory is used to study the contact conductance between two touched CNTs, and combing with the contact area and contact region number evolution information obtained via MD simulations, it could help understand the macroscopic compression influence to the CNT foam conductance.

The macroscopic conductance property of CNT foam is related to its microscopic CNT assembling style. Concretely, the twist lattice angle of touched area between two CNT is critical to their contact conductance. This is because the contact conductance between CNT forming a network in a series or parallel connection way eventually determines the CNT foam conductance. To verify this point of view, the contact area, region number and also the
bending degree evolution with respect to the compressive strain are shown in fig. S19, A and B.

The transport model is composed by two CNTs contacting with each other, which is shown in Fig. 3E where the contact area is marked with red color and the chirality of these two CNT could be different. The transport model could be divided into three regions, i.e., two leads and the middle conductance region, and electrons transport from one lead to the other. The energy resolved contact conductance is shown in Fig. 3F and fig. S19, C to F, and the twist lattice angle varies from 0 to 30 degrees, which covers all lattice twist possibilities due to the six-fold rotation and mirror symmetry. Different contact areas are set in order to study their influence, the contact area only changes the conductance amplitude, while the transport gap is gradually opened with twist lattice angle increasing. Under macroscopic compression, both the average contact area and region number between CNTs enhance. As a result, contact conductance amplitude increase, and on the other hand, the rising contact region number provides more opportunities for CNTs to touch with each other with different twist lattice angles and thus a smaller gap could be expected, which well reproduces the experimental measurement qualitatively.

One thing needs to be noted that only twist lattice angle is emphasized, which in our model is formed by choosing specific chirality of the two CNTs (it could also be formed by rotating two CNTs), since the sufficient large diameter (20 nm) of the CNT suggests a negligible curvature effect leading to a similar twist lattice angle dependent contact conductance behavior to graphene sheets.

S2.13 UV–vis diffuses absorption

According to the UV–vis diffuses absorption spectra (fig. S21A) of the CNT films, the enhanced absorbance at high strains illustrates the closely packed CNTs. As shown in fig. S21B, after irradiation under visible light for 2 minutes, the microelectrodes at 80% strain exhibit a obviously temperature enhancement, which also declared its high density of CNTs. Then, the corresponding band gap was inferred by \((A\nu)^2\) and \(\nu\) curves (fig. S21C), and the band gap decreases with the increase of strain, consistent with the results of mesoscopic transport theory (Fig. 3F and fig. S19).

S2.14 Electrochemical activity test

To investigate the electrochemical activity of CNT film at different compress strains, a comprehensive model of electrode reactions accurately describes electrochemical processes taking place on CNT electrodes for ferro-/ferricyanide electrochemicals pair. Taking into account the capacitive component, the current densities for the \([\text{Fe}^{III}(\text{CN})_6]^{3−}\leftrightarrow[\text{Fe}^{II}(\text{CN})_6]^{4−}\)
electron transfer reactions reveal a dramatic rise from 0% to 80% strain (Fig. S22a), and the peak separation decreases with increasing strain, indicating an enhanced electrochemical reactivity. In addition, the corresponding Nyquist plots (fig. S22b) can also declare the electrochemical activity of CNT film increased with the increase of strain.

**S2.15 First principles calculation**

In this paper, one of the critical issues is about hydrogen atom adhesion on CNTs since it directly correlates with the capacity of CNT foam. First principles calculation is adopted to study the adsorption energy between the hydrogen atom and graphene (the local region of CNT is approximately considered as graphene). In Fig. 3H, three possible hydrogen atom adhesion configurations are shown, where hydrogen atom position is set to top, bridge and hollow sites, respectively. The adsorption energy of $E_b=E_{Gr-H} - E_{Gr} - E_H$ are -2.11 eV, -1.37 eV and -0.433 eV for top, bridge and hollow site configuration. Obviously, the top site configuration has the lowest adsorption energy and thus it is the major site for hydrogen atom adhesion. Next, uniaxial strain along armchair and zigzag direction is applied to this configuration, the energies of graphene and graphene/H system are shown in fig. S25 for comparison. With isolate hydrogen atom energy equaling -0.0285 eV, the corresponding adsorption energy curves could be calculated (Fig. 3I). It is interesting to find out that no matter under tensile or compressive strain, the adsorption energy of Gr/H system generally decreases concluding that deformation could promote the adhesion of hydrogen atom to graphene or CNT. Under macroscopic shrinkage and wrinkling, CNT bending deformation is inevitable, and the strain distribution on the surface of CNT film is heterogeneous and could change from compressive to tensile (Fig. 3G). As both of them enhance the adhesion between the hydrogen atom and CNT, based on which the enormous capacity increment found in experiments could be understood.

**S2.16 Comparison of balloon-based MSCs and commercial battery**

Lightweight electrode design is a prerequisite for the flyable power supply system. As shown in Supplementary fig. 39A, the lightweight integrated MSCs have no effect on the balloon's ability to float normally, leaving plenty of room for appliances. On the contrary, the helium balloon in Supplementary fig. 39B wouldn’t float properly due to the heavy self-weight of commercial batteries, proving that the integrated device we designed has great application potential as a flight power supply system.
**Fig. S1. Device fabrication with homemade CNT ink (with sodium alginate binder).** The interdigital electrode on the balloon surface at (A) expansion and (B) contraction states. SEM images of crumpled CNT microelectrode with (C) low resolution and (D and E) high resolution.
Fig. S2. Characterization of CNT film. (A) SEM and (B) TEM images of CNT film. (C) XRD and (D) XPS spectra of CNT film. (E) SEM-EDS elemental analysis of CNT film.
Fig. S3. Investigation of optimum electrode material concentration. (A–E) Photographs and (F–I) SEM images of the crumpled MSCs with different concentrations of CNT ink. (J) GCD curves and (K) areal capacitance of the four devices.
Fig. S4. Tests of adhesion between CNTs and balloons. (A) Schematic diagram of the interaction between inflated balloon and CNT ink. A water droplet on the inflated balloon surface (B) before and (C) after friction treatment. (D) Water flow and (E) electroscope are used to detect charge on the surface of the balloon. (F–H) Adhesion test of CNT film on the loose plane of balloon, the surface of an inflatable balloon after friction and the surface after balloon shrinkage. (I) Inside surface of the balloon after shrinkage. (J) Adhesion strength between CNT film and balloon at 0% and 80% contraction strain (according to test criteria of GBT9286-1998).
Fig. S5. Digital images of the transformation from macro to micro supercapacitors under compress strains.
Fig. S6. Planar sizes and cross section SEM images of electrodes under different compress strains.
Fig. S7. Inference of wrinkling behavior on CNT film by Neo-Hookean bilayer model. (A) Model of CNT film on the surface of balloon under 0\% strain. (B) Maximum principal stress distribution evolution with respect to compressive strain.
Fig. S8. The nanoindentation force-depth curves of the samples.
Fig. S9. FEM calculation results of CNT structure with its substrate (the balloon) under initial 400% pre-stretched and different compression strains, the wrinkling behavior is triggered with its amplitude and periodicity gradually evolving.
Fig. S10. FEM calculation results of CNT structure with its substrate (balloon) under initial 400% pre-stretched and different compression stresses, the wrinkling behavior is triggered with its amplitude and periodicity gradually evolving.
Fig. S11. **Chemical stability of balloon.** (A) XRD and (B) CV curves of a single MSC measured at initial and after 12 h dropping the electrolyte.
Fig. S12. Electrochemical test. (A–D) CV and (E–G) GCD curves at different compress strains based on electrode volume.
Fig. S13. Capacitance enhancement efficiency versus strain curves.
Fig. S14. Slope of $Z_{\text{Re}}$ against $\omega^{-1/2}$ of the five devices.
Fig. S15. Energy/power densities and cycling stability of devices. (A) Comparation of energy/power densities of the devices at different compress strains. (B) Long-term cycling stability of the 80% strained MSC at the current density of 0.36 A cm$^{-3}$. 
Fig. S16. **Stacked MSCs.** (A–C) Digital photographs, planar sizes and cross section SEM images of stacked electrodes under different simulative compress strain.
Fig. S17. Electrochemical characterization of stacked MSCs. (A) GCD curves of the devices of stacking–20%, stacking–40% and stacking–20% measured at 0.036 A cm$^{-3}$. (B) Nyquist plots and (C) real/normalized imaginary part capacitance of stacking–80% device.
Fig. S18. MD simulation of uncovered CNT microstructure evolution under biaxial compression.
Fig. S19. Mesoscopic transport theory. (A) The evolution of average CNT bending degree with respect to compressive strain. (B) Statistics of contact area and contact regions number between different CNTs under deformation. (C--F) Evolution of the contact conductance of the lattice twist with different angle degree.
Fig. S20. SEM images. (A) SEM and (B) mapping images of CNT films at different strains.
Fig. S21. UV-Vis diffuses absorption spectra and infrared imaging. (A) UV-Vis diffuses absorption of CNT-film electrode at 0% and 80% strains. (B) Infrared thermal images of 0% and 80% strained devices with UV-Vis-NIR light illumination for 2 min. (C) (Ahv)^2 versus hv curves of the CNT films.
Fig. S22. Electrochemical activity test. (A) Typical CV curves of CNT electrodes at different strains (0%, 20%, 40% and 80%) in 12 mM K₃[Fe(CN)₆] solution. (B) Low and (C) high magnification Nyquist plots with of CNT electrodes at different strains (0, 20, 40 and 80%) in 0.5 M PBS solution with the presence of 12 mM K₃[Fe(CN)₆].
Fig. S23. Test of effective active surface area. (A–D) CV conducted at potential from 0.4 V to 0.5 V vs Ag/AgCl at scan rates of 1, 2, 5, 10, 20, 40 and 80 mV s\(^{-1}\). (A–D) The CNT films with the compress strain of 0\%, 20\%, 40\% and 80\%, respectively. (E) Capacitive current densities measured at 0.45 V vs RHE with different scan rates.
Fig. S24. Conductivity of CNT electrodes. (A) Resistance of CNT films under different strains. (B) Output current performances of CNT films at different compress strain (constant voltage of 60 mV cm⁻²). (C) Linear curve between output current and strains.
Fig. S25. Under the uniaxial strain along armchair and zigzag direction, the energies of graphene and graphene/H system are shown for comparison.
Fig. S26. Electrochemical test. (A, C, E, G) CV and (B, D, F, H) GCD curves of 80%-strain MSC based on 1M HCl, H₃PO₄, ZnSO₄ and KOH gel-electrolytes, respectively.
Fig. S27. Influence of balloon radius on device performance. (A) Initial states of the balloons with different sizes. Devices on different size of balloon with 0% (B–E) and 80% (F–M) compress strains. (N–Q) SEM images of four devices on different size of balloon with 80% compress strains. (R) GCD curves of the four devices.
Fig. S28. Rate capacities of shape-tailored MSCs at 80% compress strains.
**Fig. S29. MSC integration.** Digital images of the integrated devices connecting (A) in series and (B) in parallel at 0% and 80% strain. (C) CV profiles of integrated MSCs with 2–4 units connecting in series (s) and in parallel (p).
Fig. S30. **Shape-tailored integrated MSCs.** (A–C) Digital images of shape-tailored integrated MSCs with 0% strains. (D) CV (50 mV s⁻¹) and (E) GCD (2 μA) curves of shape-tailored integrated MSCs with 80% compress strains.
Fig. S31. Mechanical stability. (A) SEM image of CNT microelectrode after various mechanical deformations. (B) CV curves of the device before and after 200 cycles stretching and contraction at 60% strains. (C) SEM image of CNT microelectrode after 200 cycles stretching and contraction.
Fig. S32. Stretchable exhibition. (A) Schematic diagram and (B) SEM of the device at 0% and 400% stretching strain. (C) Application of stretchable MSCs.
Fig. S33. Shape adaptation of integrated devices. (A) The integrated devices in series on various 3D curvilinear substrates and (B) corresponding CV curves (50 mV s$^{-1}$). (C) Flexible MSCs embedded in the crevices of the building model, which light a string of LEDs.
Fig. S34. The integrated devices with different shapes serve as the power supply units and flexible watch chain of the electronic watch.
Fig. S35. Underwater power supply device. (A) Schematic diagram of the MSC as underwater power system. (B) CV and (c) GCD curves of the underwater MSC with artificial seawater electrolyte.
Fig. S36. Spherical mask and integrated devices. (A, B) Design sketch and (C) photograph of the mask. (D) Balloon inflated inside the mask was sprayed with CNT ink. (E) Scaled prepared devices on the balloon through the spherical mask. (F) Integration strategy of the large scaled devices.
Fig. S37. Application of integrated devices. (A) Integrated devices light a series of LEDs. (B) The digital graph of the integrated devices and lighted LEDs on the surface of helium balloon.
Fig. S38. Electrochemical performance test at different temperatures. The device tests were performed at (A) room temperature, (B) 0 °C, and (C) -10 °C. (D) CV curves measured at different temperatures.
Fig. S39. Comparison of balloon based MSCs and commercial battery. The helium balloons binding with integrated devices (A) and commercial batteries (B).
Table S1. Areal capacitance and energy density comparison of different CNT-based devices.

| Electrodes | Electrolytes          | Scan rates | Specific capacitances | Energy densities | Ref. |
|------------|-----------------------|------------|-----------------------|------------------|------|
| CNT        | PVA-H$_3$PO$_4$       | 20 µA cm$^{-2}$ | 2.44 mF cm$^{-2}$    | -                | 20   |
| CNT        | [EMIM][TFSI] ionic liquid | 6 µA cm$^{-2}$ | 0.51 mF cm$^{-2}$    | 0.034 µWh cm$^{-2}$ | 21   |
| CNT        | Ionic liquid gel      | 0.1 V s$^{-1}$ | 0.43 mF cm$^{-2}$    | 0.1~0.5 µWh cm$^{-2}$ | 22   |
| CNT        | PVA-LiCl              | -          | 1.68 mF cm$^{-2}$    | -                | 23   |
| CNT        | LiPF$_6$              | 1 mV s$^{-1}$ | 1.0 mF cm$^{-2}$    | -                | 24   |
| G/CNT      | Na$_2$SO$_4$          | 200 mA cm$^{-2}$ | 2.16 mF cm$^{-2}$ | -                | 25   |
| C/chitosan-coated CNT | H$_2$SO$_4$          | 10 mV s$^{-1}$ | 6.09 mF cm$^{-2}$ | -                | 26   |
| MnO$_2$/CNT | PVA-H$_3$PO$_4$      | 2 µA       | 3.01 mF cm$^{-2}$    | -                | 27   |
| rGO/CNT    | KCl                   | 10 mV s$^{-1}$ | 6.1 mF cm$^{-2}$    | -                | 28   |
| CNT        | PVA-H$_2$SO$_4$       | 50 µA cm$^{-2}$ | 13.5 mF cm$^{-2}$ | 1.4 µWh cm$^{-2}$ | This work |
Table S2. Volume-specific capacitance and energy density comparison of electrodes in different CNT-based MSCs.

| Electrodes   | Electrolytes | Scan rates | Specific capacitances | Energy densities | Ref. |
|--------------|--------------|------------|-----------------------|------------------|------|
| CNT          | PVA-H₃PO₄    | 10 mV s⁻¹  | 2.02 F cm⁻³           | 0.09 mWh cm⁻³    | 31   |
| CNT          | PVA-H₃PO₄    | 0.05 V s⁻¹ | 1.86 F cm⁻³           | -                | 29   |
| CNT          | PVA-H₃PO₄    | 0.009 A cm⁻³ | 5.0 F cm⁻³      | 0.4 mWh cm⁻³    | 30   |
| CNT/GO       | PVA-H₃PO₄    | 1 A cm⁻³   | 3.1 F cm⁻³           | 0.84 mWh cm⁻³    | 32   |
| CNT/rGO      | KCl          | 1 V s⁻¹    | 5 F cm⁻³             | 0.68 mWh cm⁻³    | 28   |
| CNT/agarose  | PVA-H₃PO₄    | 7.7 mA cm⁻³ | 1.2 F cm⁻³     | -                | 33   |
| CNT/AgNW     | PVA-H₃PO₄    | 10 mV s⁻¹  | 2.29 F cm⁻³          | 0.17 mWh cm⁻³    | 34   |
| CNT          | PVA-H₂SO₄    | 0.036 A cm⁻³ | 9.3 F cm⁻³   | 0.99 mWh cm⁻³    | This work |
Table S3. Stretchable degree comparison of various MSCs and MBs (micro-batteries).

| Devices                  | Electrode patterns | Elastic substrates         | Stretched direction | Stretched ratio | Ref. |
|--------------------------|--------------------|-----------------------------|---------------------|-----------------|------|
| Ti$_3$C$_2$Tx Mxene MSC  | 3D interdigital    | PDMS                        | Uniaxial            | 50%             | 49   |
| Mn/Mo oxide-CNTs MSC     | 2D interdigital    | PDMS/Ecoflex                | Biaxial             | 50%             | 50   |
| MWCNT/PANI MSC           | 3D interdigital    | PDMS                        | Uniaxial            | 40%             | 52   |
| rGO MSC                  | 2D interdigital    | PDMS                        | Uniaxial            | 60%             | 53   |
| rGO MSC                  | 2D interdigital    | PDMS                        | Uniaxial            | 100%            | 51   |
| Graphene/NiO/Co$_3$O$_4$ | 2D interdigital    | Waterborne polyurethane     | Uniaxial            | 100%            | 54   |
| MSC                      | 2D interdigital    | Rubber                      | Uniaxial            | 100%            | 55   |
| PEDOT:PSS/Graphene MSC   | 2D interdigital    | Rubber                      | Uniaxial            | 100%            | 56   |
| Zinc-ion MB              | 3D interdigital    | Ecoflex                     | Uniaxial            | 40%             | 57   |
| Zinc-ion MB              | 3D interdigital    | Fibroid                     | Uniaxial            | 50%             | 58   |
| CNT-based MSC            | 2D interdigital    | Balloon                     | Omnidirectional     | 400%            | This work |
Movie S1: Surface charge detection of an expansion balloon.
Movie S2: Fully charged integrated devices light up the watch.
Movie S3: Fully charged device array light up a string of LEDs.
Movie S4: Exhibition of a flyable power supply system.