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Supporting Information

Ultrahighly Sensitive and Repeatable Flexible Pressure Sensor Based on the MWCNTs/PVDF/PU Hierarchical Frameworks-structured Aerogels for Monitoring Human Activities

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Experimental section

Preparation of the PPMs. PPMs was prepared by a facile solvothermal method. First, polyVinylideneFluoride (PVDF) was added in the dimethyl formamide (DMF) solvent to form the 10% PVDF/DMF solution. Then, different MWCNTs were added in the 10% PVDF/DMF solution and shook for about 2h to form mixed solution with doping different MWCNTs. And then, 2ml PU prepolymer was added in 4 ml MWCNTs/PVDF/DMF mixed solution accompanied with shocking. The wet gels with PVDF/PU framework was instantly generated, where MWCNTs was uniformly distributed. Furthermore, the obtained material was placed in plasma vacuum processing 30min and then dried directly at 70°C for 1h in a vacuum oven, resulting in slight increase in volume. To further decrease the density, the bulked PPMs monolith was heated at 250 °C in a vacuum oven over night. Finally, the PPMs with a density of 13.5mg/cm^3 was obtained. It was obvious that the fabricated process was fundamentally distinguished from other reported complicated drying approaches of fabrication of carbon aerogels, e.g., freeze drying and supercritical drying, thus suitable to large-scale production.

Characterizations. The morphology of the as-prepared samples was investigated using the scanning electron microscope (Hitachi S-4800). X-ray photoelectron spectra were recorded on a ULVAC-PHI 5000 Versa Probe II. Elements analysis and mapping by an environmental scanning electron microscope (Quanta 250 FEG). Raman spectra were recorded on a Horiba

LABRAM HR Raman spectrometer with an excitation wavelength of 633 nm. Fourier transform infrared spectra were recorded on a Nicolet iN10 spectrometer. Electric properties were measured by using a Keithley 4200 SourceMeter controlled by a LabVIEW based data acquisition system at room temperature. Mechanical property was performed by Intron 3365. Thermogravimetric analysis was performed on a Seiko DSC6220 instrument.

Preparation and measurement of pressure sensor:The pressure sensor was constructed based on the PDMS substrate and the PPMs film. To improve the contact between the PPMs film and the substrate, the film of PPMs based pressure sensor was fabricated using coating method. The mixed solution with 3% MWCNTs and 10% PVDF in the DMF solvent was firstly coated on the treated PDMS substrate, and then the PU pre-polymer was subsequently coated, where the volume ratio of the MWCNTs/PVDF to PU pre-polymer was 2:1. Instantly, the wet gel with PVDF/PU framework was generated. The obtained gel was treated using the same method as the above mentioned description. Well-cut PPMs (15mm×7mm×0.4mm) were connected to aluminium wires with silver paint. The computer-controlled Keithley 4200 SourceMeter was used to record the current variation, and the pressure was applied on the upper surface.

Preparation and Measurement of compressive strain sensor: The compressive strain sensor was constructed based on sandwich structure. The active layer was a cylindrical shape (the bottom radius R=1cm, height H=1.5cm) PPMs aerogels.

Two pieces of silver pastes with similar shape were used as the electrodes of the sensor. Compressive tests were carried out using a Mark-10 instrument equipped with two flat-surface compression stages. The current response during the compression was recorded in real time using a Keithley 4200 Source Meter controlled by a Labview based data acquisition system. The applied voltage was 4V and all measurements were measured in the atmosphere. The output voltage was obtained from the oscilloscope system (Wavepro 760Zi-A).

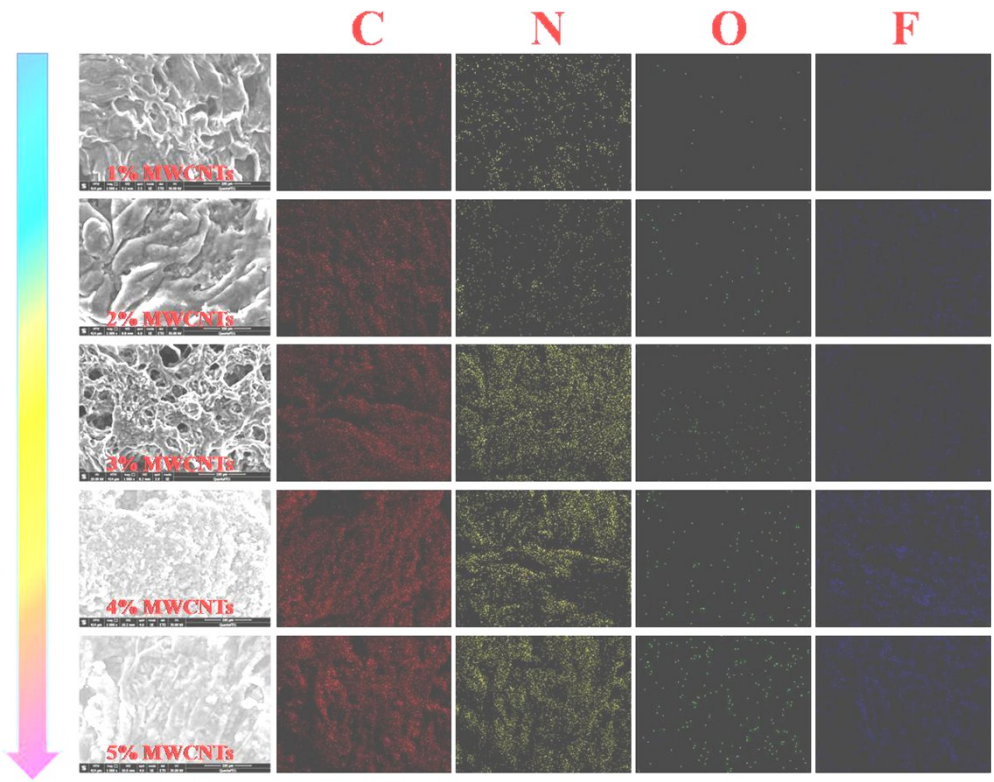

Figure S1. The element mapping of the PPMs with different doping MWCNTs.

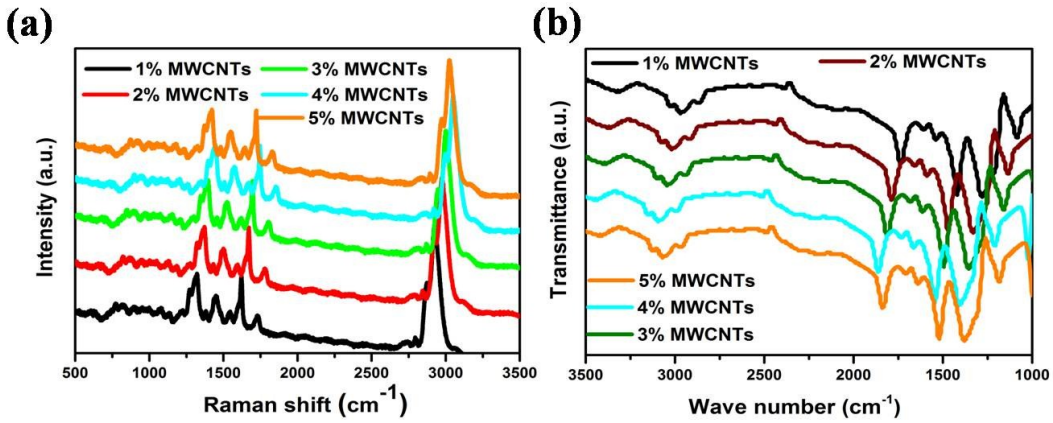

Figure S2. The Raman spectroscopy and the FT-IR spectra of PPMs with different doping MWCNTs.

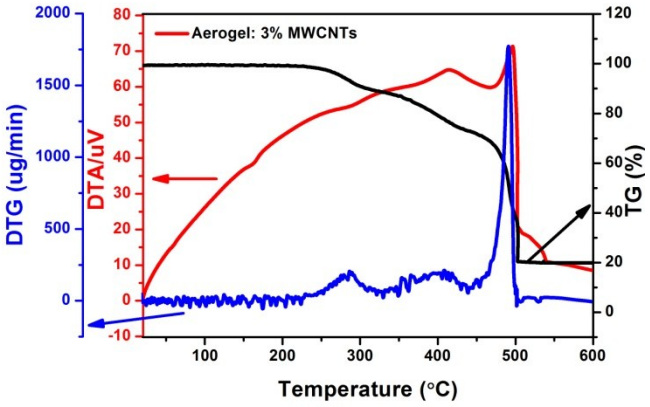

Figure S3. The thermogravimetric (TG) and the differential thermal gravimetric analysis (DTG) curves of the PPMs

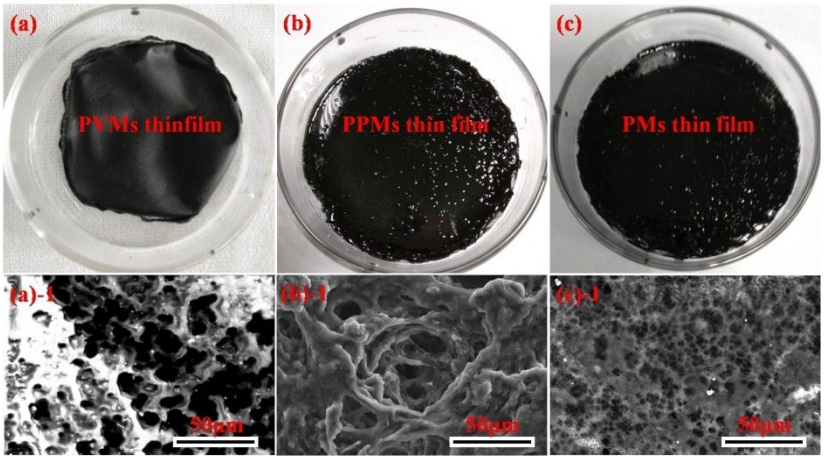

Figure S4. The photograph and the SEM of different aerogels based on different polymer. PVDF/PU/MWCNTs(PPMs), PVDF/MWCNTs (PVMs),and PU/MWCNTs (PMs).

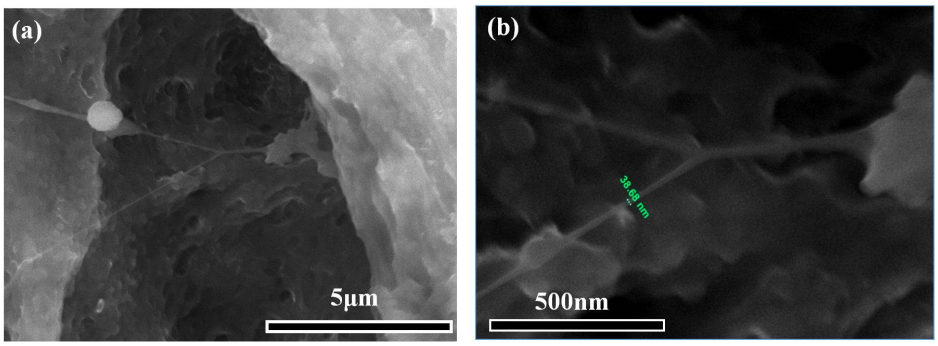

Figure S5. The magnified SEM of PPMs.

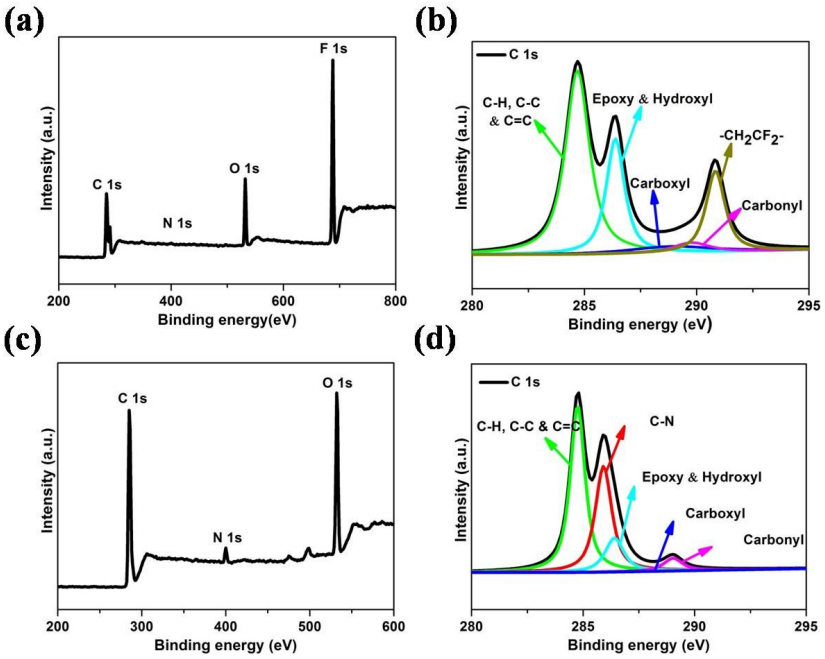

Figure S6. The XPS of the (a-b) PVDF/MWCNTs (PVMs). (c-d) PU/MWCNTs (PMs).

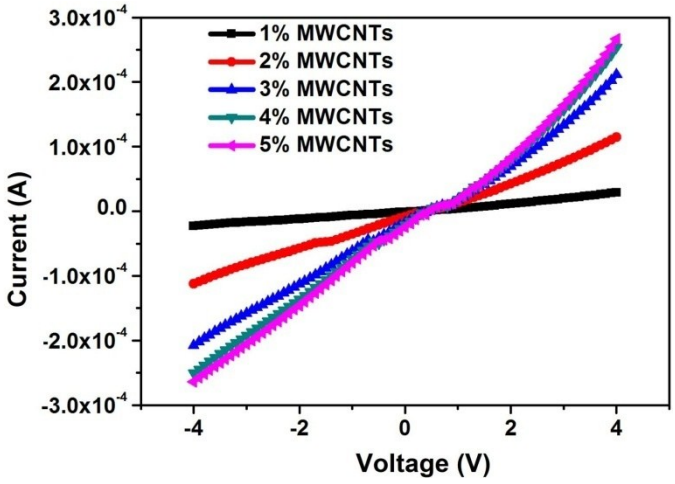

Figure S7. The current-voltage curves of the PPMs aerogels with different doping MWCNTs.

Figure S8. The instrument of the measuring the mechanical properties.

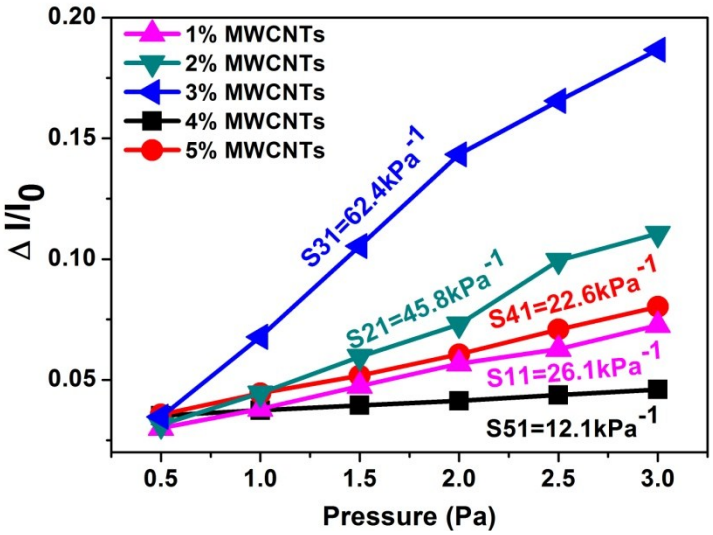

Figure S9. The relative current change under small pressures at the bias voltage of 4V.

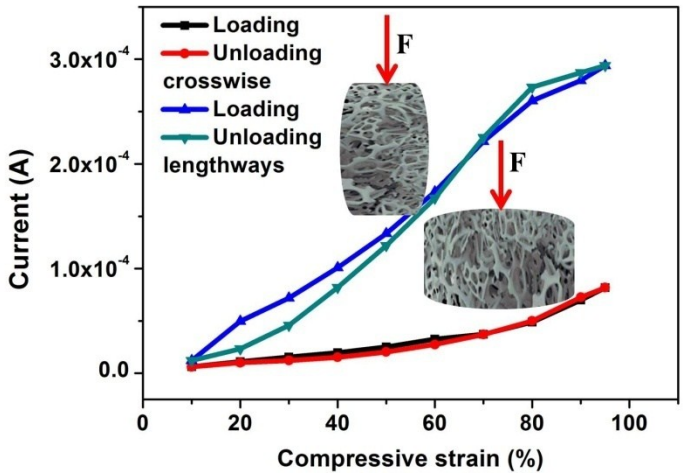

Figure S10. The current-strain curves of the PPMs aerogels when loading different directions.

Table S1 Comparison the performances of pressure or compressive sensors based on reported CNT aerogels materials

Sample	Density (mg/cm^3)	Sensitivity (kPa^{-1})	Detection limit (Pa)	Strain range (ε/%)	Strain sensitivity	References
1	5-10	/	/	80	/	[33]
2	240	/	/	/	/	S1
3	2.52			80		[34]
4	/	0.2	0.5	/	/	[29]
5	10-54	/	/	70-95	0.24	[35]
6	5.8-25.5	/	/	60	0.32	S2
7	14	/	/	80	0.5	[28]
8	4-6	/	/	80	0.94	S3
9	/	/	/	60	15	S4
10	/	/	/	/	134	[30]
11	/	/	/	15	/	[39]
12	7.5-37		6	60	0.9	S5
This work	13.5	62.4	0.15	95	156.4	*

S References

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