

Electronic Supplementary Information

**An intrinsically compressible and stretchable all-in-one configured  
supercapacitor**

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## Experimental Part

### Preparation of the PAA hydrogel electrolyte.

AA (36g , Macklin) was first added into de-ionized water (36 mL) under vigorous stirring in 0°C ice bath until the solution was uniformly mixed. 10 min later, APS (0.585g, Thermo Fisher) was added into the solution. After the solution was stirred in 0°C ice bath for 30 min, the solution was injected into our mold and polymerized in an oven at 45°C for 30 hours.

### Preparation and electrochemical characterization of the all-in-one configured supercapacitor.

CNTs (0.061 g, Suzhou TANFENG Graphene Tech Co, Ltd) and H<sub>3</sub>PO<sub>4</sub> (6.1 g, Macklin) were added into de-ionized water (24.4 mL) at room temperature. Then the solution was sonicated until CNTs were evenly dispersed. Trace amount of pyrrole (1.5 mL, Thermo Fisher) was dropped into the solution under vigorous stirring in 0°C ice bath. Another mixed solution of APS (0.73 g, Thermo Fisher), H<sub>3</sub>PO<sub>4</sub> (4.1 g, Macklin) and de-ionized water (16.4 mL) was blended thoroughly into the first solution. An as-synthesized PAA hydrogel was cut into the size of 2 cm\*3 cm, and then immersed into the blended solution for 3 min. Then the four edges were cut to avoid the short circuit and the all-in-one configured supercapacitor was obtained.

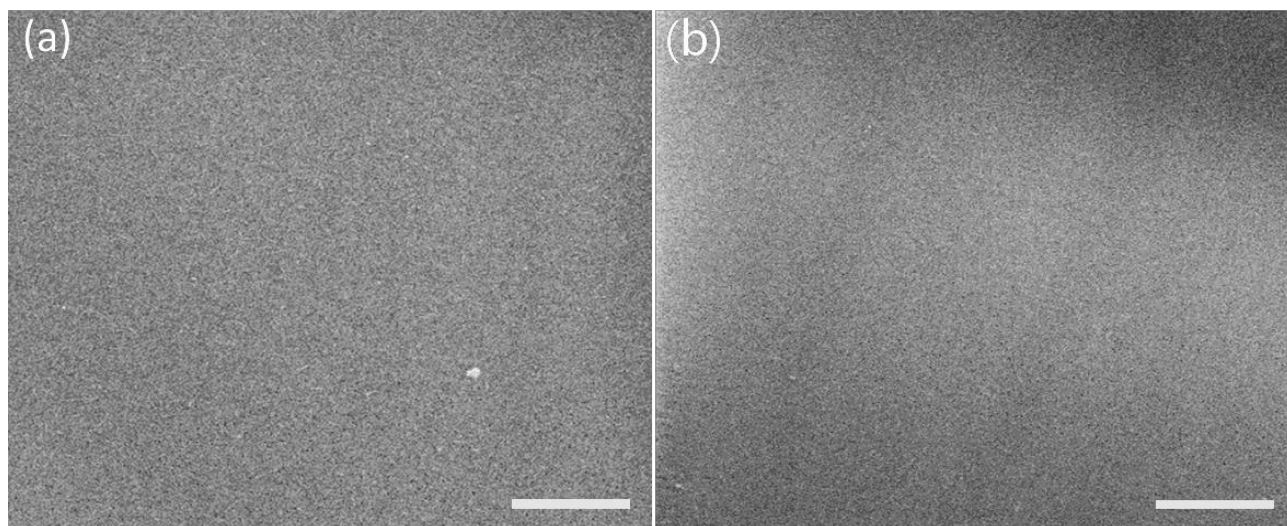
The electrochemical performance of the all-in-one configured supercapacitor was measured by CV and GCD in a two-electrode configuration using CHI760E potentiostat. All measurements were performed at room temperature. The capacitance with respect to the single electrode ( $C_s$ ) was calculated using the charge integrated from GCD and CV curves individually according to the following formulas:

$$C_s = \frac{2It}{U_s} \quad (1)$$

$$C_s = \frac{1}{U_{vs}} \int_{U_-}^{U_+} i(U) dU \quad (2)$$

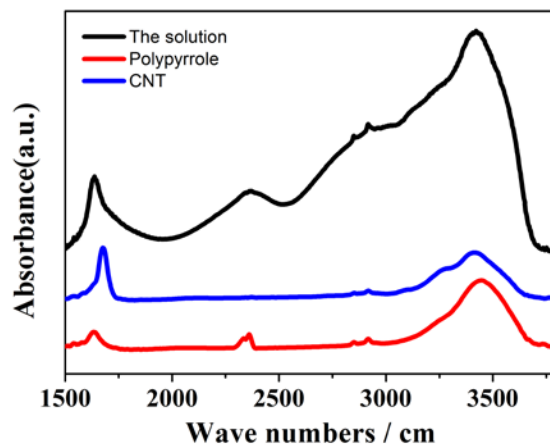
where  $I$  is the discharge current during GCD,  $t$  is the discharge time during GCD,  $U$  is the voltage range ( $U = U_+ - U_-$ ),  $s$  is the area of one electrode,  $v$  is the scan rate of the CV curve, and  $i(U)$  is the current during CV.

## Figures



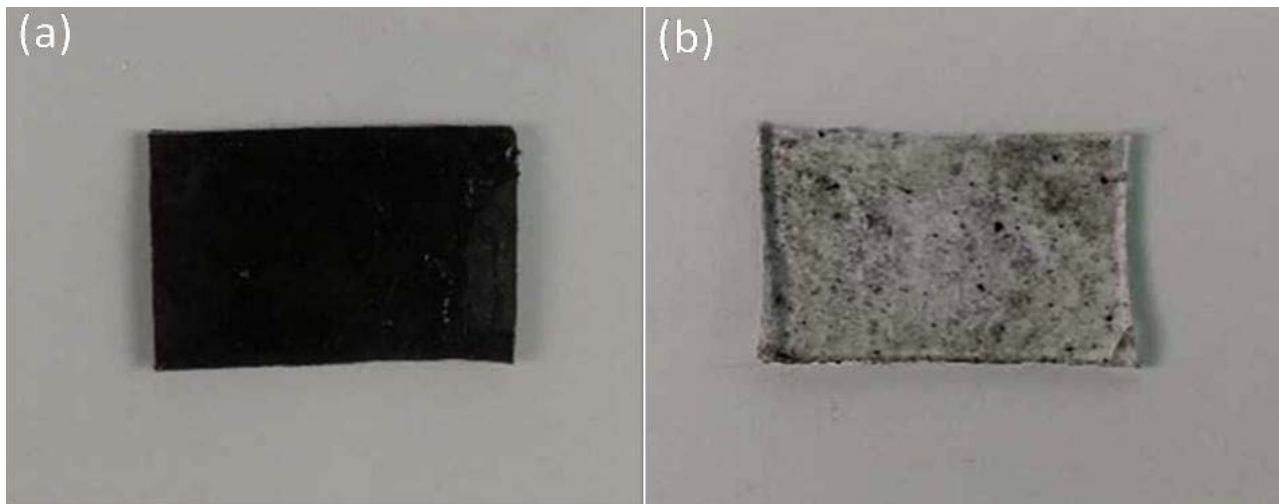
**Fig. S1. Characterization of the PAA hydrogel before and after stretch.** (a) An SEM image of the PAA hydrogel before stretch. Scale bar: 1  $\mu\text{m}$ . (b) An SEM image of the PAA hydrogel after stretch. Scale bar: 1  $\mu\text{m}$ .

The original PAA surface is non-directional. After being stretched, it appears some fringes along the stretch direction without any microscopic cracking and breakage on the surface. This signifies that the molecular chains are stretched, as schematically represented in Figure 2b in the manuscript.



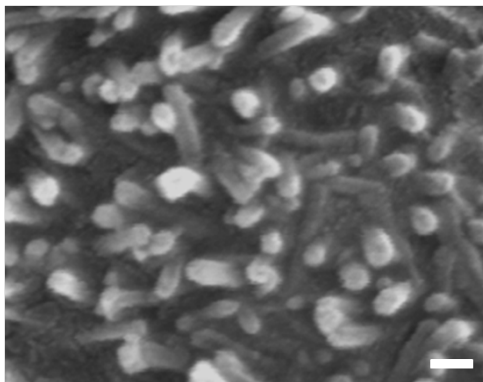
**Fig. S2. FTIR spectra of the solution, polypyrrole and CNT.**

The characteristic peaks of the solution coincide with those of polypyrrole and carbon nanotube, which prove that the solution contains polypyrrole and carbon nanotube.



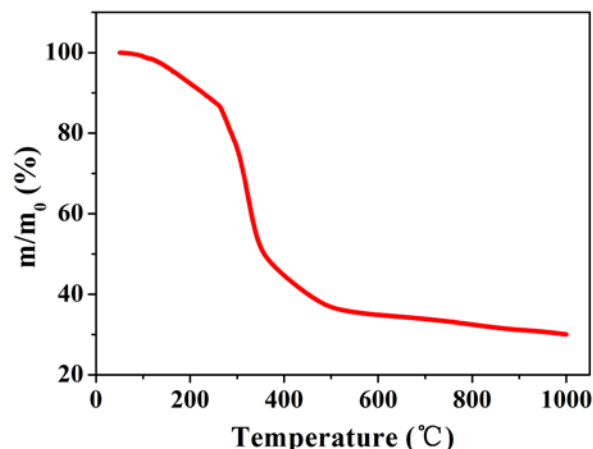
**Fig. S3. Preparation of the all-in-one configured supercapacitor in different solutions.** (a) The all-in-one configured supercapacitor fabricated in the solution with the pyrrole. (b) The all-in-one configured supercapacitor fabricated in the solution without pyrrole.

Pyrrole is very important for fabricating the all-in-one configured supercapacitor. Without the pyrrole in the solution, CNTs can not be attached on the electrolyte on a large scale which results in the failure of fabricating the all-in-one supercapacitor.



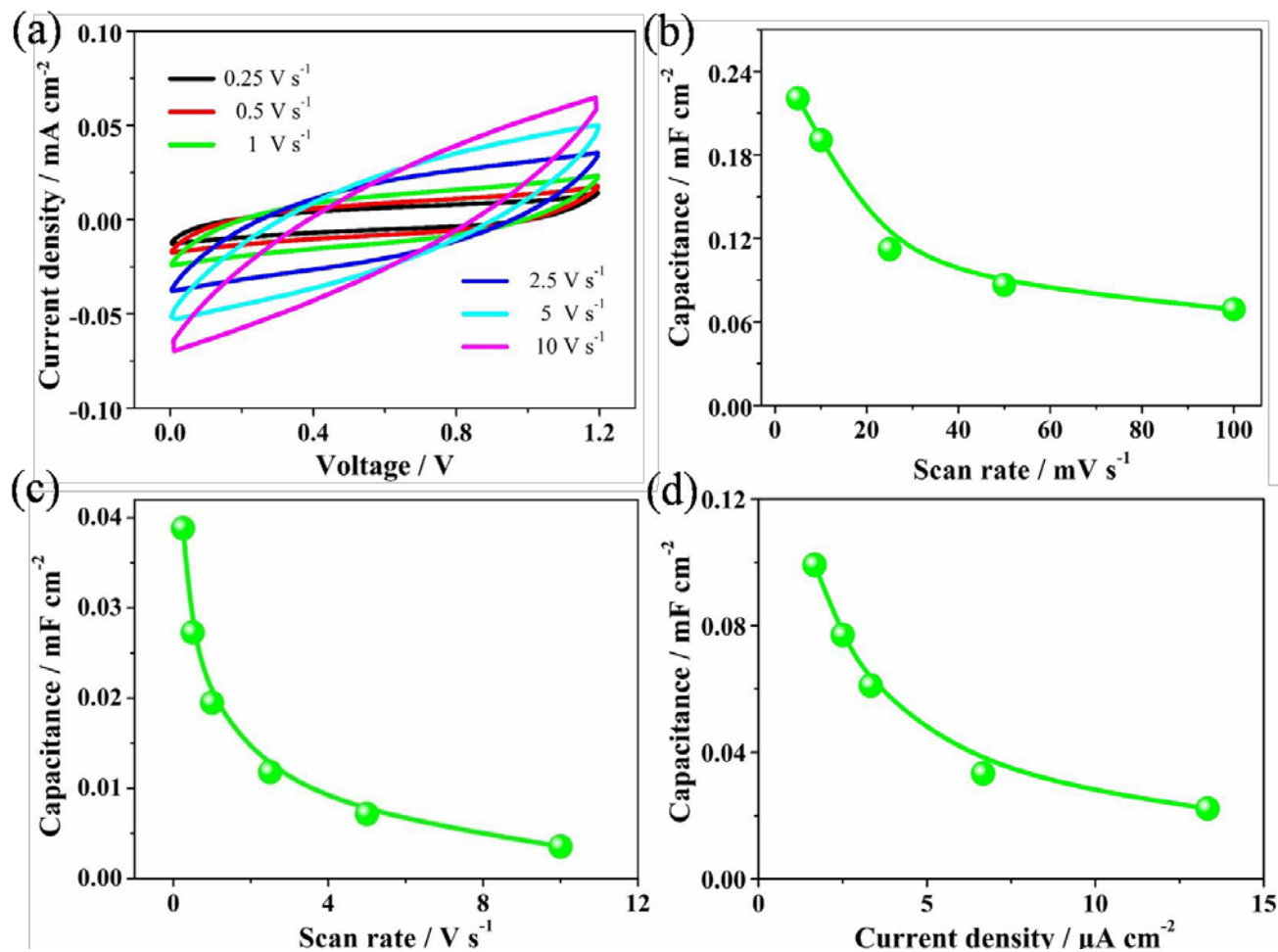
**Fig. S4. An SEM image of CNTs on the surface of the all-in-one configured supercapacitor.  
Scale bar: 50 nm.**

Carbon nanotubes were remarkably observed on the all-in-one configured supercapacitor.



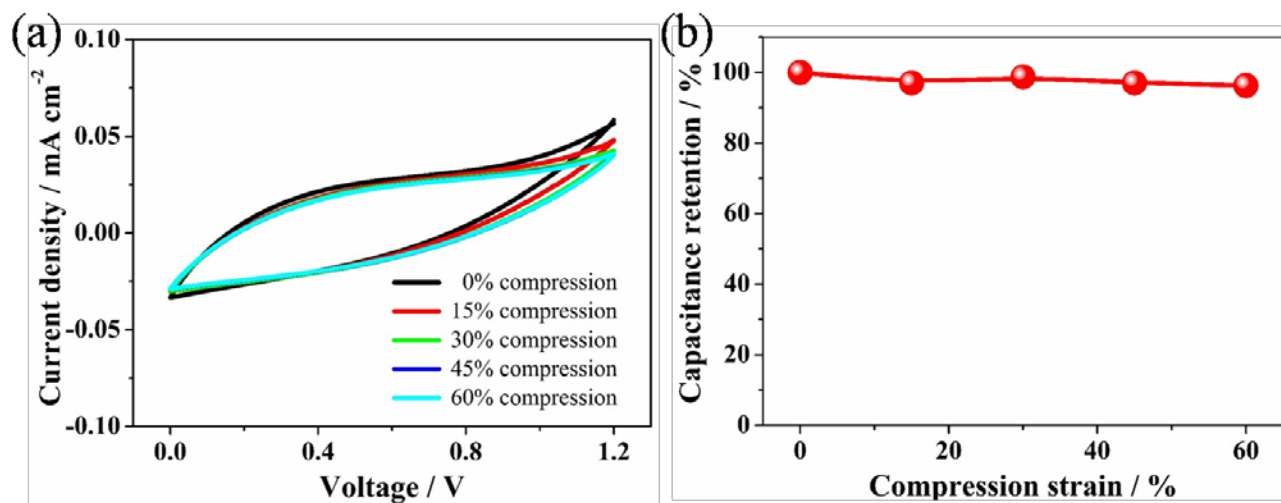
**Fig. S5. TGA curve of the all-in-one configured supercapacitor.**

In the first stage, it lost water. In the second stage, the decomposition of -COOH in PAA and of N-H in PPy was occurred. Finally, the total carbon content was about 30 wt. %. In order to accurately measure the loading of carbon nanotubes, we prepared two solutions. The first solution was prepared according to our experimental method. The second solution was the same as the first solution except that carbon nanotubes were not added. Two pieces of the PAA with the same size were respectively placed in these two solutions with identical time, and then were taken out and weighed at both wet and dry state. Based on the mass difference between them, the loading of carbon nanotubes was confirmed to be 3.4 wt. %.



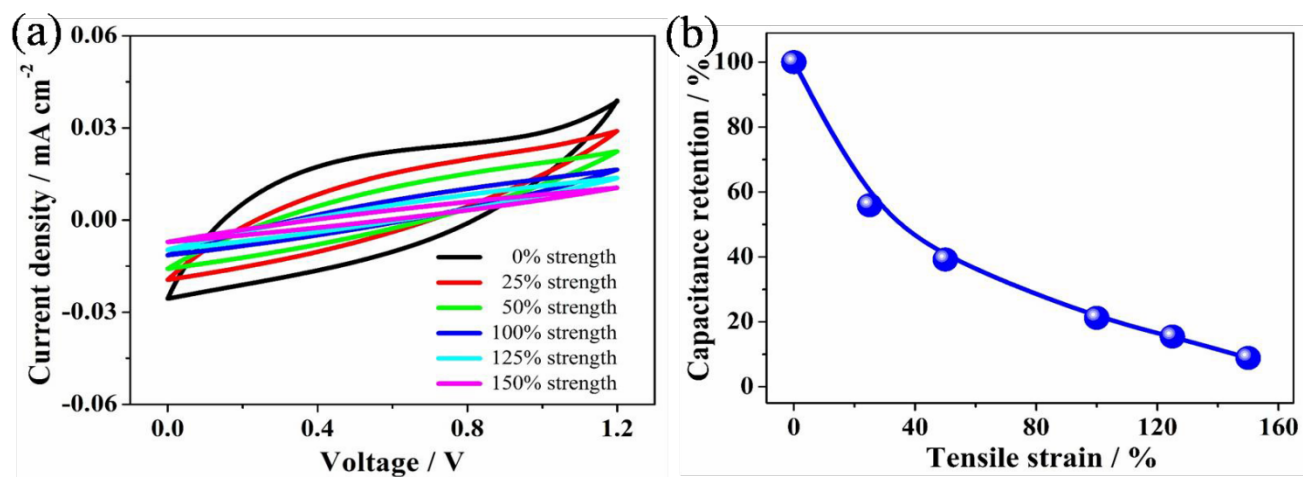
**Fig. S6** Electrochemical performance of the all-in-one configured supercapacitor. (a) CV curves at various scan rates from 0.25 to 10  $\text{V s}^{-1}$ . (b), The specific capacitance of the all-in-one configured supercapacitor as a function of scan rates from 5 to 100  $\text{mV s}^{-1}$ . (c) The specific capacitance of the all-in-one configured supercapacitor as a function of scan rates from 0.25 to 10  $\text{V s}^{-1}$ . (d) The specific capacitance of the all-in-one configured supercapacitor as a function of specific current density from 1.7 to 13  $\mu\text{A cm}^{-2}$ .

The cyclic voltammetry (CV) curves at scan rates up to 10  $\text{V s}^{-1}$  indicate that the supercapacitor can endure very fast voltage/current change. With the increase of scan rate, CVs gradually deviate from the rectangular shape and the capacitance decreases. Similarly, the capacitance calculated from GCD curves also decreases with the increase of the current density.



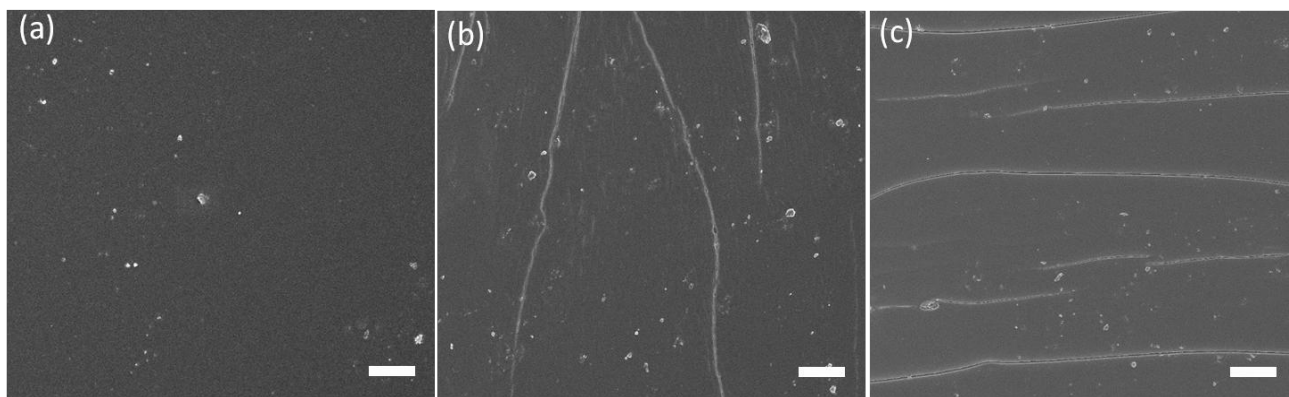
**Fig. S7.** Electrochemical performance of the all-in-one configured supercapacitor under compression. (a) CV curves from 0-60% compression at 100 mV/ s. (b) Capacitance retention obtained from CV curves as a function of the compressive strain.

The all-in-one configured supercapacitor can achieve 60% compression with the electrochemical performance still retains well at various compressive strains.



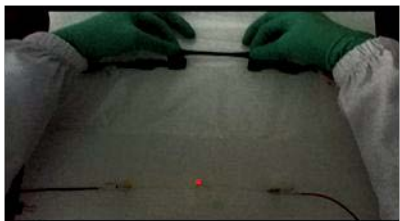
**Fig. S8.** Electrochemical performance of the all-in-one configured supercapacitor under stretching. (a) CV curves from 0-150% strength at 100 mV s<sup>-1</sup>. (b) Capacitance retention obtained from CV curves as a function of the strength strain.

The all-in-one configured supercapacitor can be stretched to 150% with the electrochemical performance still existed. The capacitance retention calculated from CVs shows a tendency of decrease with the increase of stretch.



**Fig. S9. Characterization of the all-in-one configured supercapacitor under stretching.** (a) An SEM image of the relaxed supercapacitor. (b) An SEM image of the 50% stretched supercapacitor. (c) An SEM image of the 125% stretched supercapacitor. All scale bars: 10  $\mu\text{m}$ .

The decline of the capacitance during the stretch results from the enlarged crack on the electrode surface.



**Fig. S10.** All-in-one configured supercapacitors power a LED bulb during stretch.

The all-in-one configured supercapacitors can light the LED bulb for over 30 seconds during repeated stretch.