Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supplementary information:

Highly Stretchable and Flexible Supercapacitors Based on Electrospun PEDOT:SSEBS Electrodes

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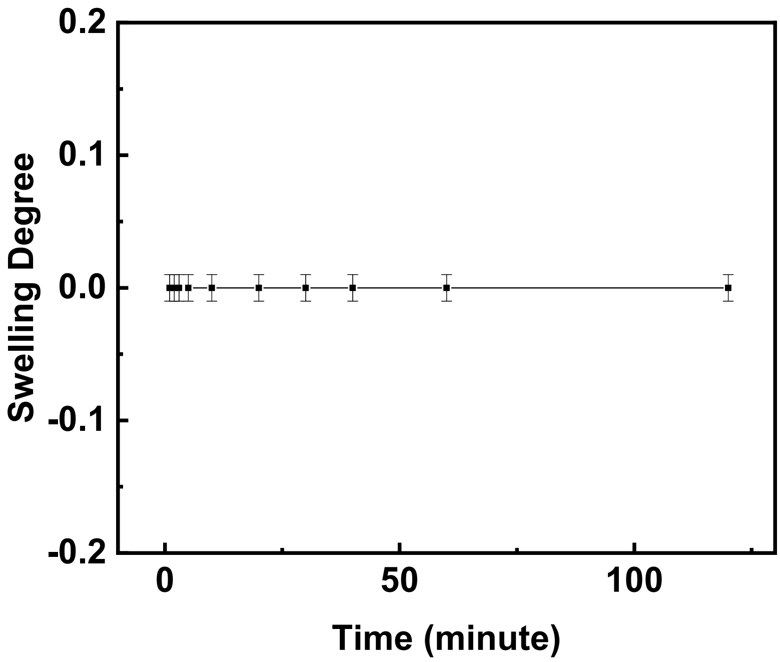

Fig. S1 Swelling test for SSEBS fibrous mat conducted in methanol for different times.

Raman Spectroscopy

Cross sectional samples for Raman spectroscopy analysis were prepared by snap freezing microfiber mat samples in liquid nitrogen and breaking the samples in half to create a cross sectional edge. The samples were compressed between two glass coverslips and mounted under the microscope objective turret. The Samples were imaged using the LabSpec6 viewsharp mosaic function which adjusts the stage z position for optimal camera focus and stitches the fields of view with 20% overlap (Figure S2).

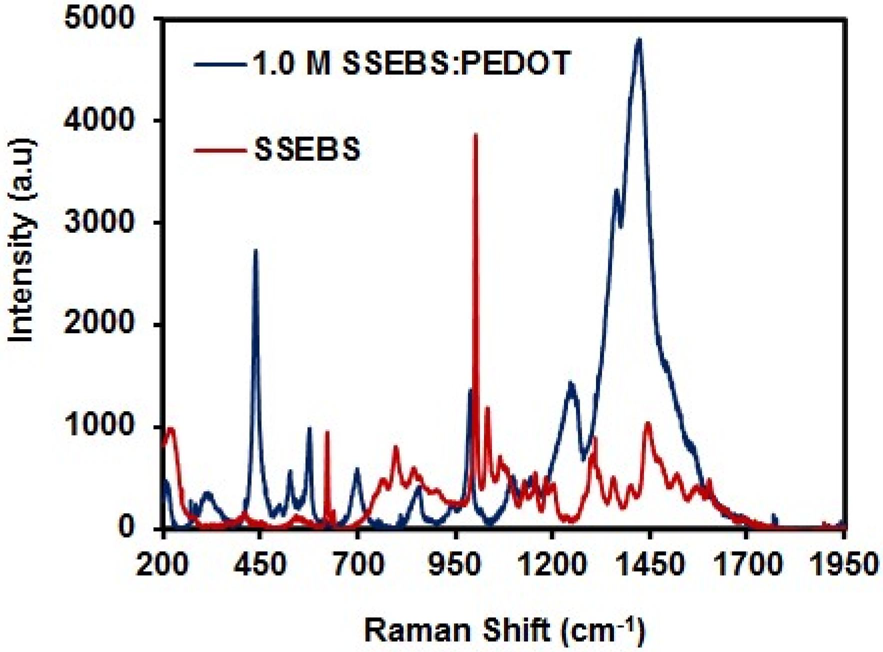

Fig. S2 Bulk measured Raman spectra of 1.0 M PEDOT:SSEBS and control SSEBS microfiber mats.

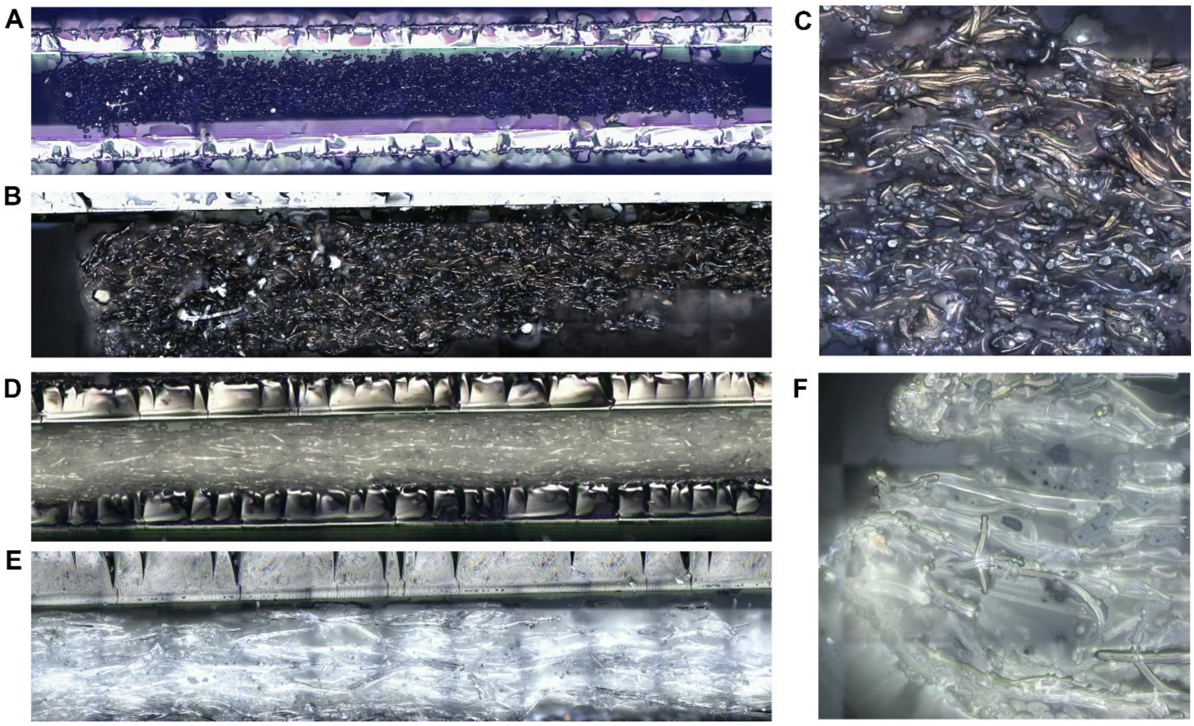

Fig. S3 Viewsharp mosaic images of cross-sectional samples of 1.0 M PEDOT at A: 10x magnification, B: 50x magnification and C: 100x magnification and SSEBS control sample at D: 10x magnification, E: 50x magnification and F: 100x magnification.

For all Raman data acquisitions the 785 nm excitation was passed through a neutral density filter that allows 1% transmission of the Rayleigh line to the objective lens. This produced a laser spot with a diameter of approximately 5 µm.

Bulk measurements of the SSEBS:PEDOT and SSEBS control sample (Figure. S1) were measured using a wide field confocal hole (200 µm). The SSEBS control sample required 120 s exposure time and 25% laser power to achieve adequate signal strength, while the PEDOT infused sample provided strong singles with 60 s exposure time and 1% laser power. The confocal pinhole acts as a spatial filter, by allowing the Raman spectrometer to look into a smaller spatial domain than with a conventional configuration without the pinhole, attenuating the out of-focus regions of the sample. A line map across the diameter of the microfiber cross section was measured with 0.5 µm steps in the x direction on 3 microfibers (Figure. S3). This data was collected using a 50 µm confocal hole. This provided a 0.532 µm theoretical resolution in x, y by the Rayleigh Criterion equation (Equation. S1)

$$x,y\ plane\ (lateral){:}d = 0.61\,\frac{\lambda}{N.A} \qquad \textbf{Equation. S1}$$

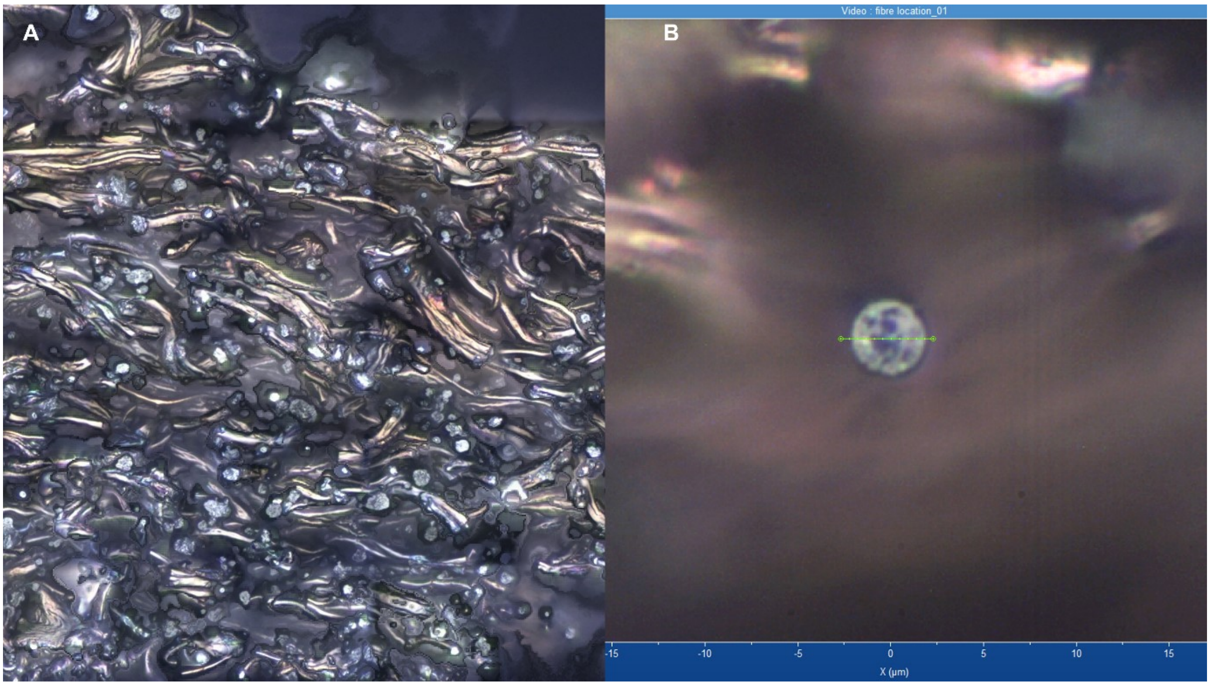

Fig. S4 (A) Viewsharp mosaic image of 1.0 M PEDOT infused SSEBs sample at 100x magnification. (B) Microfiber cross-section line map coordinates with 0.5 µm step sizes.

All Raman Data was baseline corrected using Labspec 6.5 software (Horiba Japan). The spectra were fitted with a third order polynoimial baseline with 130 anchor points. The baseline subtracted spectra were smoothed using a Savitsky-Golay algorithm with a third order polynomial and a 5 point symmetric kernel in Aspen Unscrambler V12.2. The spatially resolved line map data set was area normalised and the peak height at 1425 cm^{-1} was determined.

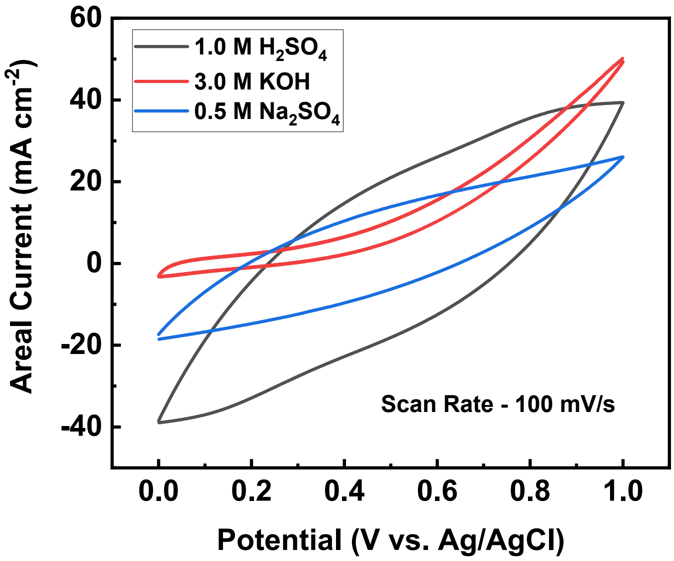

Fig. S5 CVs recorded at 100 mV s^{-1} of 1.0 M PEDOT:SSEBS microfiber mat in different aqueous electrolytes: 1.0 M H_2SO_4, 3.0 M KOH and 0.5 M Na_2SO_4.

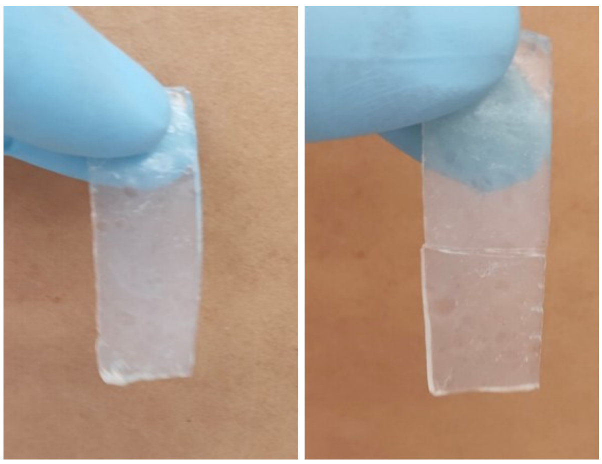

Fig. S6 Self-healing property of the PVA-H_2SO_4 gel electrolyte. A cut gel strip self-healed within 20 s.

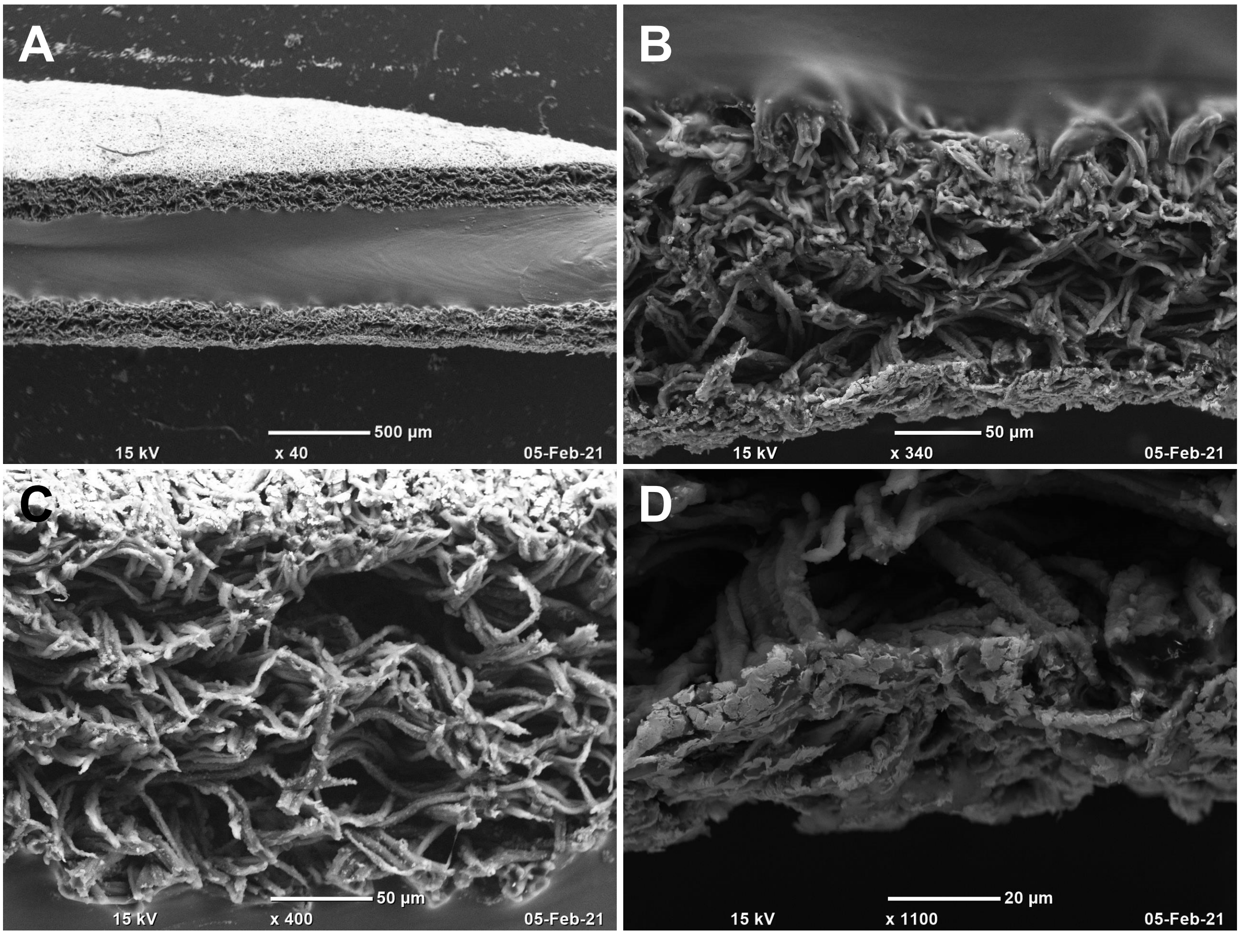

Fig. S7 (A) Cross-sectional SEM images of the stretchable supercapacitor of thickness 0.6 mm, constructed from 0.5 M PEDOT:SSEBS microfiber mats on top and bottom, and PVA-H_2SO_4 gel electrolyte in the middle. Thickness of the PVA-H_2SO_4 gel electrolyte was approximately 500 µm. (B)-(D) Enlarged cross-sectional SEM images of the PEDOT:SSEBS microfiber mat at the bottom.

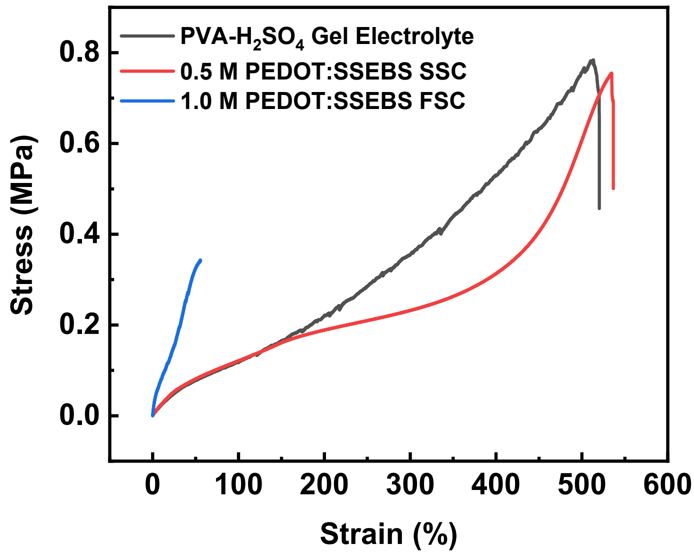

Fig. S8 Stress-strain curves of the PVA-H_2SO_4 gel electrolyte, stretchable supercapacitor constructed from 0.5 M PEDOT:SSEBS microfiber mats and PVA-H_2SO_4 gel electrolyte, and bendable supercapacitor constructed from 1.0 M PEDOT:SSEBS microfiber mats and PVA-H_2SO_4 gel electrolyte.

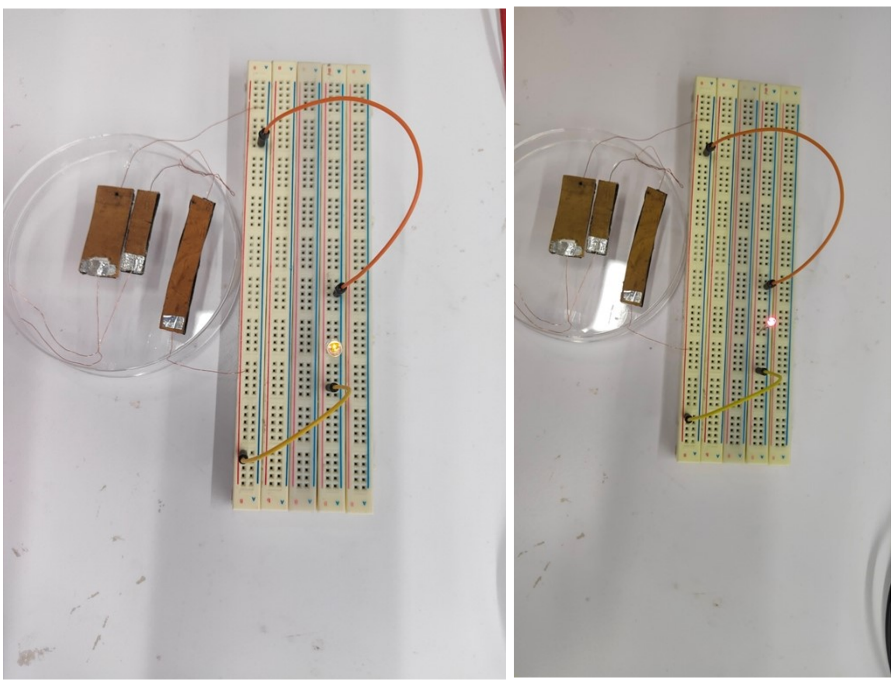

Fig. S9 Photographic image of the LED lighting using 0.5 M PEDOT:SSEBS microfiber mat and PVA-H_2SO_4 gel electrolyte based stretchable supercapacitor (yellow (left) and red (right) LED). Three devices were connected in series.

Table S1. Fitting parameters of Nyquist plots before and after 15,000 charge discharge cycles of the stretchable supercapacitor device constructed from 0.5 M PEDOT:SSEBS microfiber mats and PVA-H_2SO_4 gel electrolyte.

	R_S (Ω)	Q_1 ($F.s^{(a_1-1)}$)	a_1	Q_2 ($F.s^{(a_2-1)}$)	a_2	R_{CT} (Ω)	R_d (Ω)
Before 15,000 cycles	5.771	0.0803	1	0.229	1	1.452	59.4
After 15,000 cycles	10.22	0.062	0.7261	0.08	0.946	0.043	0.877

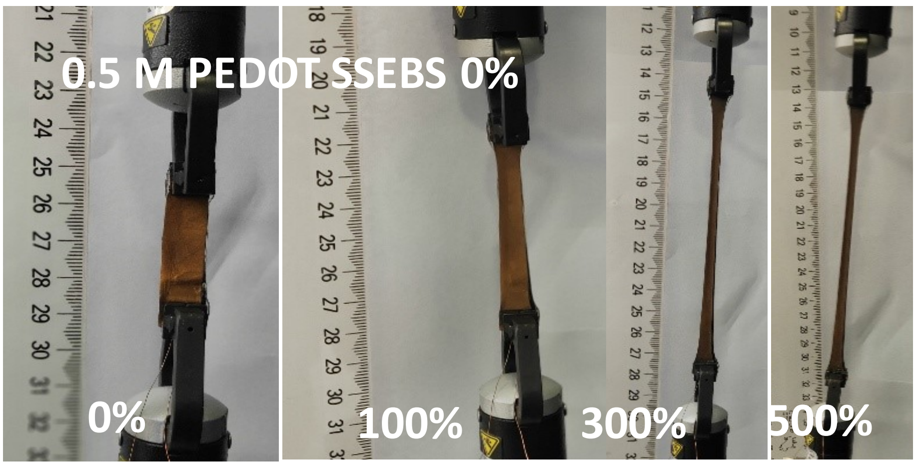

Fig. S10 Stretchablilty test of the supercapacitor device constructed from 0.5 M PEDOT:SSEBS microfiber mats and PVA-H_2SO_4 gel electrolyte, showing elongations up to 500%.

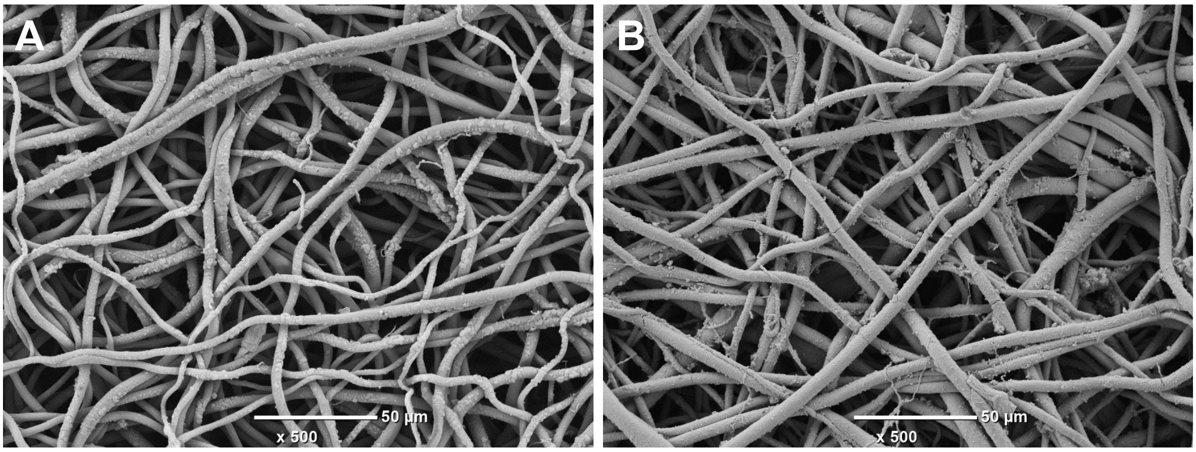

Fig. S11 SEM images of the stretchable device before (A) and after stretching up to the breaking point (B).

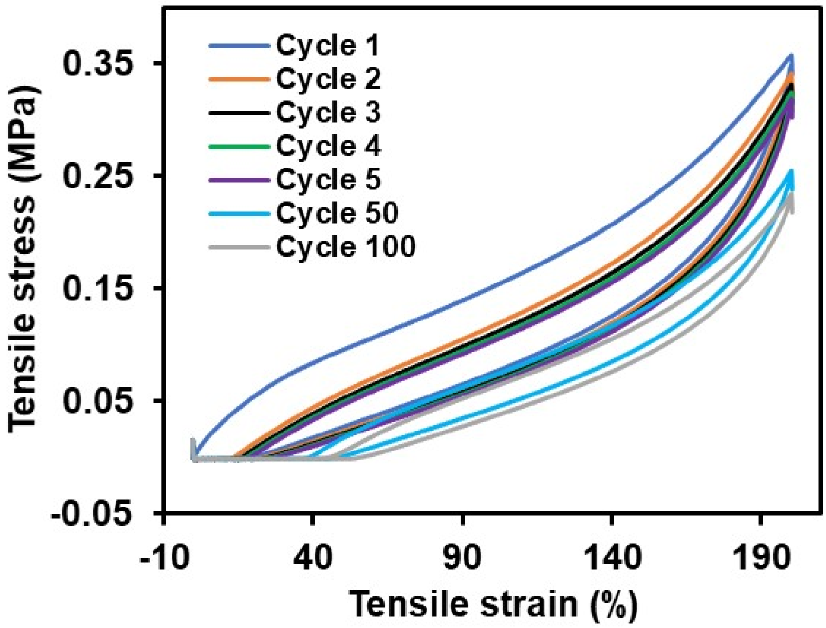

Fig. S12 Cyclic tensile stress–strain curves of the stretchable device; strain rate was 10%/s.

Table S2 Comparison of electrochemical performances of the 1.0 M PEDOT:SSEBS microfiber mat electrode in 1.0 M H_2SO_4 with PEDOT based electrodes recently reported in the literature.

Electrode material	Electrolyte	Potential window	Areal capacitance	Ref.
PEDOT:PSS films	Human sweat	0.8 V	10 mF cm^{-2} at 1 mV s^{-1}	1
Graphene-PEDOT/PSS films	1.0 H_3PO_4	1.0 V	448 mF cm^{-2} at 10 mV s^{-1}	2
PEDOT array on carbon fibers	1.0 M H_2SO_4	1.0 V	184 F g^{-1}	3
Ag-doped PEDOT: PSS/CNT composites	1.0 H_3PO_4	0.8 V	64 mF cm^{-2}	4
CNTs/PEDOT sponge	1.0 M H_2SO_4	0.8 V	147 F g^{-1}	5
MnO_2-graphene oxide-PEDOT:PSS nanocomposite	0.5 M Na_2SO_4	1.0 V	177 mF cm^{-2}	6
1.0 M PEDOT:SSEBS microfiber mat	1.0 M H_2SO_4	1.0 V	1396 mF cm^{-2}	Current work

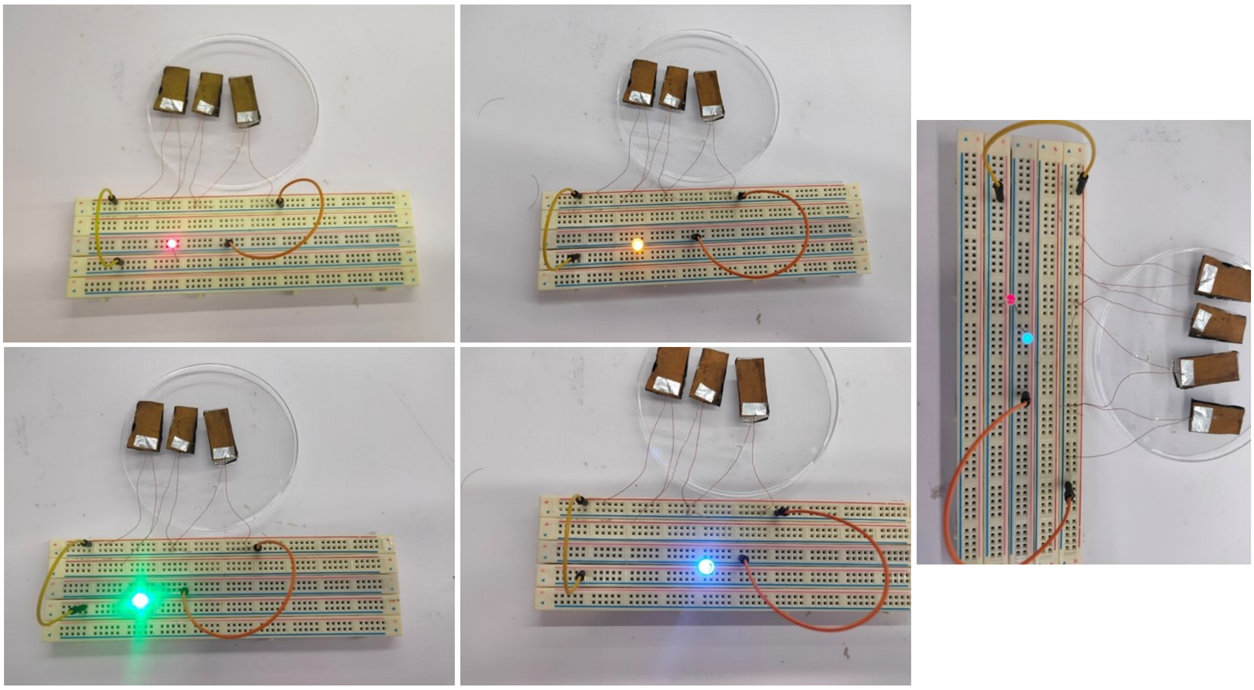

Fig. S13 Photographic image of the LED lighting using 1.0 M PEDOT:SSEBS microfiber mat and PVA-H_2SO_4 gel electrolyte based bendable supercapacitor. Three devices were connected in series.

Table S3 Fitting parameters of Nyquist plots before and after 5,000 charge discharge cycles of the flexible supercapacitor device constructed from 1.0 PEDOT:SSEBS microfiber mats and PVA-H_2SO_4 gel electrolyte.

	R_S (Ω)	Q_1 ($F.s^{(a_1-1)}$)	a_1	Q_2 ($F.s^{(a_2-1)}$)	a_2	R_{CT} (Ω)
Before 5000 cycles	2.22	0.434 9	0.891 4	0.1676	0.642	3.238
After 5000 cycles	3.628	0.2923	0.9148	0.1421	0.5214	9.385

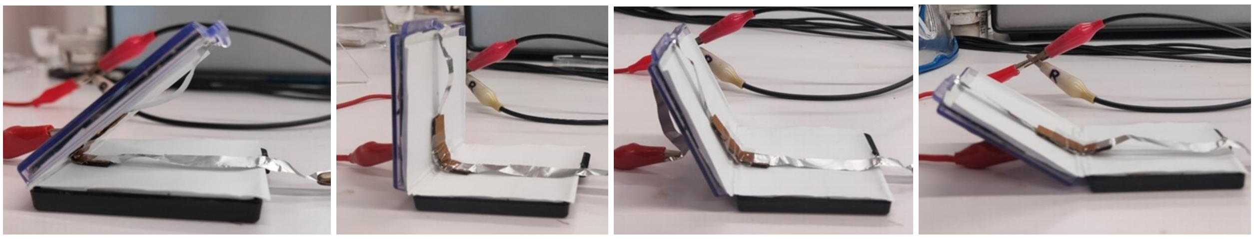

Fig. S14 Bending test conducted at different bending angles using the solid state supercapacitor constructed from 1.0 M PEDOT:SSEBS microfiber mat and PVA-H_2SO_4 gel electrolyte.

Fig. S15 Stretchability test of the supercapacitor device constructed from 1.0 M PEDOT:SSEBS microfiber mats and PVA-H_2SO_4 gel electrolyte, showing break starts at 60% of elongation.

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