# A colour-tunable, weavable fibre-shaped polymer light-emitting electrochemical cell

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#### **Supplementary Movie**

The word of "A" made from fibre-shaped polymer light-emitting electrochemical cells (PLECs) was selectively lightened for different parts.

#### **Supplementary Methods**

**Materials.** Stainless steel wires were purchased from Alfa Aesar. Zinc acetate dihydrate  $(Zn(CH_3COO)_2 \cdot 2H_2O, 99.999\%)$ , ethanolamine  $(NH_2CH_2CH_2OH, 99.5\%)$ , 2-methoxyethanol (CH<sub>3</sub>OCH<sub>2</sub>CH<sub>2</sub>OH, 99.8%), PEO (M<sub>V</sub> of ~100,000), THF (Tetrahydrofuran, anhydrous, 99.9%, inhibitor-free), trimethylolpropane ethoxylate triacrylate (ETT-15) and LiTf (99.995%) were obtained from Sigma-Aldrich. The soluble PF-B (polyfluorene copolymer) and SuperYellow (phenyl substituted poly(1,4-phenylene vinylene)) were obtained from 1-Materials.

Synthesis of spinnable carbon nanotube (CNT) arrays. CNT arrays were synthesized by chemical vapor deposition with Fe (1.2 nm)/Al<sub>2</sub>O<sub>3</sub> (3 nm) on a silicon substrate as the catalyst at 740 °C. For Fe/Al<sub>2</sub>O<sub>3</sub> catalyst, Fe and Al<sub>2</sub>O<sub>3</sub> were successively deposited on silicon substrate by electron beam evaporation with rates of 0.5 and 2 Å s<sup>-1</sup>, respectively. Ethylene was used as the carbon source, and a gas mixture of Ar and H<sub>2</sub> was used as the carrier gas. The flow rates of Ar, H<sub>2</sub>, and C<sub>2</sub>H<sub>4</sub> were typically 400, 30, and 90 sccm, respectively. CNT arrays with a thicknesses of 220 µm were mainly used in this work.

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Wrapping CNT sheet onto the modified steel wire. Figure 1b schematically shows the aligned CNT sheet being wrapped onto a modified stainless steel wire. The two ends of the modified stainless steel wire are firstly connected with two motors, and a spinnable CNT array is fixed onto a precisely motorized translation stage. An aligned CNT sheet (thickness of 18 nm) was then continuously drawn out of the spinnable CNT array and attached onto the modified fiber with an angle of  $\alpha$ . The thickness of the stacked CNT sheet on the modified stainless steel wire can be controlled by varying the helical angle and width of the CNT sheet.

**Calculations of the thicknesses of CNT sheet on the modified steel wire.** During the wrapping process of CNT sheet, the angle between modified steel wire and CNT sheet was 30°, which had been maintained when the precession velocity of the CNT sheet was equal to the moving velocity (*v*) of the translation stage. During the fabrication time (*t*), the area of the used CNT sheet (*S*<sub>1</sub>) can be calculated by  $S_1=v\times t\times a/\cos 30^\circ$ , and the surface area of the midified steel wire (*S*<sub>2</sub>) can be calculated by  $S_2=2\pi r\times v\times t$ . Here *a* and *r* correspond to the width of the CNT sheet (1.5 mm) and radius of the steel wire, respectively. The layer numbers of CNT sheets (*n*) were calculated by  $n=S_1/S_2$ . Typically, the layer number of CNT sheet was calculated to be 1.08 and the thickness of the stacked CNT sheet was calculated to be ~19 nm (Supplementary Table S1).

Calculation of luminance of the fibre-shaped PLEC. The fibre-shaped PLEC can

be illuminated for 360 degrees, so it is difficult to simulate such a situation. At the same time, the measured spot size of the PR650 is larger than the effective area of the fibre-shaped PLEC. Therefore, the actual luminance (*L*) of the fibre-shaped PLEC can be calculated by the product of the measured luminance (*L*<sub>0</sub>) and the ratio (*n*) between area of the measured spot size and effective area. The detailed calculation is described below (Supplementary Fig. S25). The radius (*R*) of the measured spot size was equal to 1.50 mm and the area of the spot *S*=1.92 mm<sup>2</sup>. A stainless steel wire with a diameter (*d*) of 510 µm was used as substrate for the fibre-shaped PLEC, so the radius (*a*) of the stainless steel wire was 0.255 mm. As  $\cos \alpha = a/R$ ,  $\alpha$  can be calculated to be 70.1°. As *b*=*R*sin $\alpha$ , *b*=0.705 mm. For the area of the triangle *S*<sub>1</sub>=*a*×*b*/2, *S*<sub>1</sub>=0.09 mm<sup>2</sup>. For the fan-shaped area *S*<sub>2</sub>= $\pi \times \alpha \propto R^2/360$ , *S*<sub>2</sub>=0.344 mm<sup>2</sup>. As *S*<sub>3</sub>=*S*<sub>2</sub>-*S*<sub>1</sub>, *S*<sub>3</sub>=0.254. For the effective area, *S*<sub>4</sub>=*S*-4*S*<sub>3</sub>, so *S*<sub>4</sub>=0.904 mm<sup>2</sup>. The ratio (*n*) between the area of the measured spot size and effective area can be calculated by *n*=*S*/*S*<sub>4</sub>=2.12. Therefore, the actual luminance can be calculated by *L*=*n*×*L*<sub>0</sub>.

**Characterization.** The structures were characterized by scanning electron microscopy (Hitachi FE-SEM S-4800 operated at 1 kV). The current-voltage-light intensity curves of fibre-shaped PLECs were measured with a Keithley 2400 source meter and a Photoresearch PR-650 (measurement spot size, 1.5 mm) by increasing the applied voltage from 0 to 13 V in 500 mV incremental steps. All PLEC measurements were carried out in the glove-box filled with dried argon at room temperature. Transmittance spectra were recorded by a Shimadzu UV-2550 spectrophotometer. The light emission turn-on response curves of the fibre-shaped PLECs were recorded by a Keithley 2400 source meter and a calibrated silicon photodetector under voltage pulse

between 0 and 11 V. All measurements were made in a glovebox filled with dried argon unless specified otherwise.

Table S1. Parameters for wrapping aligned CNT sheets onto the modified steel wire

with different diameters.

Diameter of the metal wire (µm)	127	510
Wrapping angle of CNT sheet (°)	15	30
Width of the CNT sheet (mm)	1	1.5
Thickness of the stacked CNT sheet (nm)	23	19



Figure S1. Chemical structures of PF-B, ETT-15 and LiTf in the emissive layer.



Figure S2. High resolution transmission electron microscopy image of a CNT.



**Figure S3.** Scanning electron microscopy (SEM) image of a spinnable CNT array at a low magnification.



Figure S4. SEM image of a spinnable CNT array at a high magnification.



Figure S5. Photographs of a CNT-wrapped polymer fibre wrapped with aligned CNT

sheet being bent into various shapes.



Figure S6. Dependence of electrical resistance of the CNT-wrapped polymer fibre on bent times. R and  $R_0$  corresponding to the resistance before and after bending, respectively.



**Figure S7.** Dependence of optical transmittance on wavelength for an aligned CNT sheet (thickness of 18 nm).



Figure S8. Photograph of an aligned CNT sheet on a labelled paper.



Figure S9. SEM image of a bare stainless steel wire at a low magnification.



Figure S10. SEM image of a bare stainless steel wire at a high magnification.



Figure S11. ZnO nanoparticles coated on the stainless steel wire.



Figure S12. Electroluminescent polymer layer coated on the ZnO nanoparticle layer.



Figure S13. Wrapped CNT sheet at a high magnification.



Figure S14. Photograph of a fibre-shaped PLEC.



Figure S15. Photographs of the flexible fibre-shaped PLEC being wrapped onto a glass rod (a), under bending (b).



Figure S16. Lifetime test of a fibre-shaped PLEC driven at 30 mA cm<sup>-2</sup>.



**Figure S17.** Polarized electroluminescence spectra of an emissive polymer film containing PF-B, ETT-15 and LiTf (weight ratios of 20:10:1) (inserted, CIE 1931 x–y chromaticity diagram showing a blue colour of the PF-B).



**Figure S18.** Photographs of a blue fibre-shaped PLEC with a diameter of 127  $\mu$ m biased at increasing voltages of 6 (a), 7 (b), 8 (c) to 9 (d) V.



**Figure S19.** Polarized electroluminescence spectra of an emissive polymer film containing SuperYellow, PEO, ETT-15 and LiTf (weight ratios of 20:2:2:1) (inserted, CIE 1931 x–y chromaticity diagram showing a yellow colour of the SuperYellow). The inserted photograph shows a fibre-shaped PLEC biased at 9 V.



Figure S20. Photograph of a metal wire with alternated fibre-shaped PLECs (biased

at 9 V).



**Figure S21.** Dependence of luminance on bent cycle. Here  $L_0$  and L correspond to the luminance before and after bending, respectively. The inserted photograph shows a PLEC with a diameter of 127  $\mu$ m biased at 10 V.



Figure S22. Surface characterization of the polymer and aligned CNT sheet. a and b. SEM images of the polymer layer before and after bending, respectively. c and d. SEM images of the aligned CNT layer before and after bending, respectively.



Figure S23. Photographs of flexible clothes woven with fibre-shaped PLECs.



**Figure S24.** Photographs of woven fibre-shaped PLECs being selectively lightened (biased at 9 V).



Figure S25. Schematic illustration to the calculation of the actual luminance.