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

Supplementary Information

Experimental Section

Materials

PEO (M_V of ~100,000), tetrahydrofuran (anhydrous, 99.9%, inhibitor-free), trimethylolpropane ethoxylate triacrylate (ETT-15) and LiTf (99.995%) were obtained from Sigma-Aldrich. The insulating fiber was purchased with the insulating polymer (e.g., polyester) coated on the Cu wire. 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

Spinnable CNT arrays were synthesized by chemical vapor deposition using Fe/Al_2O_3 as the catalyst at 740 °C. For the Fe/Al_2O_3 catalyst, Al_2O_3 (3 nm) and Fe (1.2 nm) were consequently deposited on silicon substrate by electron beam evaporation with rates of 2 and 0.5 Å/s, respectively.[S1,S2] C_2H_4 and a gas mixture of Ar and H_2 were used as the carbon source and the carrier gas, respectively. The gas flow rates of Ar, C_2H_4 and H_2 were 400, 90, and 30 cm^3/min, respectively.

Wrapping aligned carbon nanotube (CNT) sheet onto the fiber substrate

Fig. S1 schematically shows the aligned CNT sheet being wrapped onto a fiber substrate.[S2] The two ends of the fiber are fixed by 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 fiber with an angle of α. The thickness of the stacked CNT sheet on the fiber substrate can be exactly controlled by varying the helical angle and width of the CNT sheet.

Calculations of the thicknesses of outer CNT sheet wrapped onto the modified fiber

During the wrapping process of CNT sheet, the angle between modified fiber and CNT sheet was 30º, 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_1) can be calculated by $S_1=v\times t\times a/\cos 30^\circ$, and the surface area of the midified steel wire (S_2) 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 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 1.13 and the thickness of the stacked CNT sheet was calculated to be ~20 nm.

Characterization

Transmittance spectra were recorded by a Shimadzu UV-2550 spectrophotometer with the aligned CNT sheet (5×5 mm^2) paved onto the glass. The structures were characterized by scanning electron microscopy (Hitachi FE-SEM S-4800 operated at 1 kV). The current-voltage-light intensity curves were measured by a Keithley 2400 source meter and a Photoresearch PR-680 by increasing the applied voltage from 8 to 30 V in a 1 V incremental step. The light emission turn-on response curves of the fiber-shaped PLECs were measured by a Keithley 2400 source meter and a calibrated silicon photodetector under voltage pulse between 0 and 21.5 V. All PLEC measurements were carried out at room temperature.

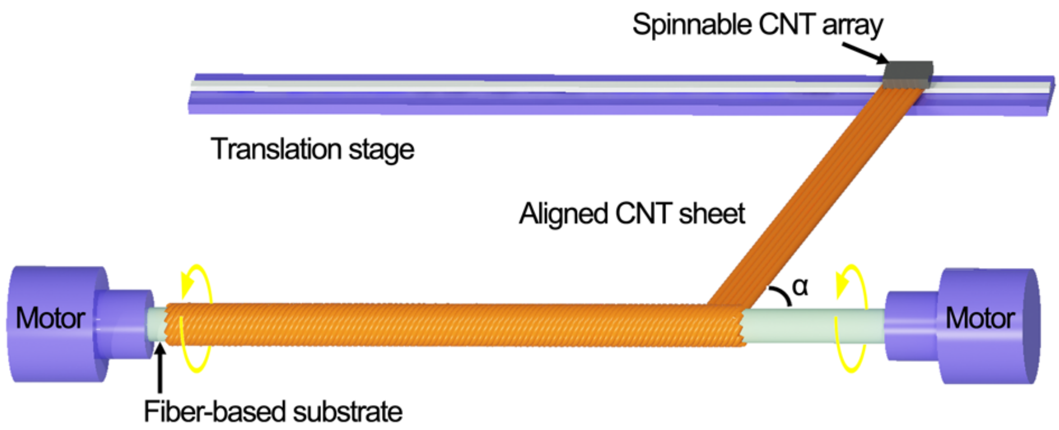

Fig. S1 Schematic illustration to the process of wrapping aligned CNT sheet onto a fiber substrate.

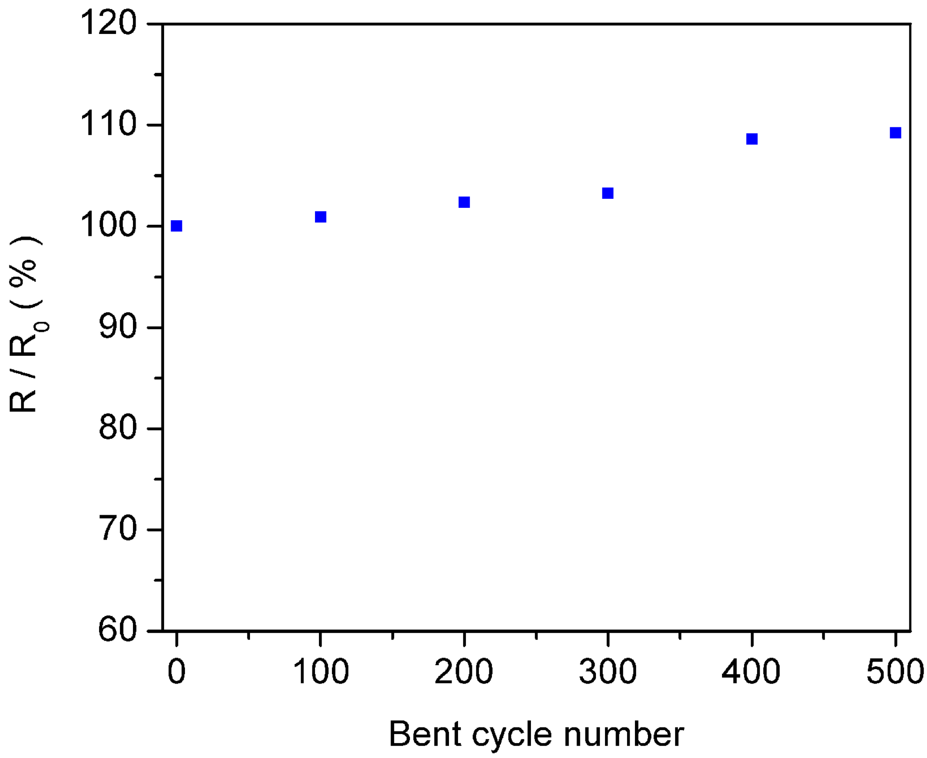

Fig. S2 Dependence of resistance variation of conducting fiber substrate on bent cycle number. R and R_0 correspond to the electrical resistances before and after bending, respectively.

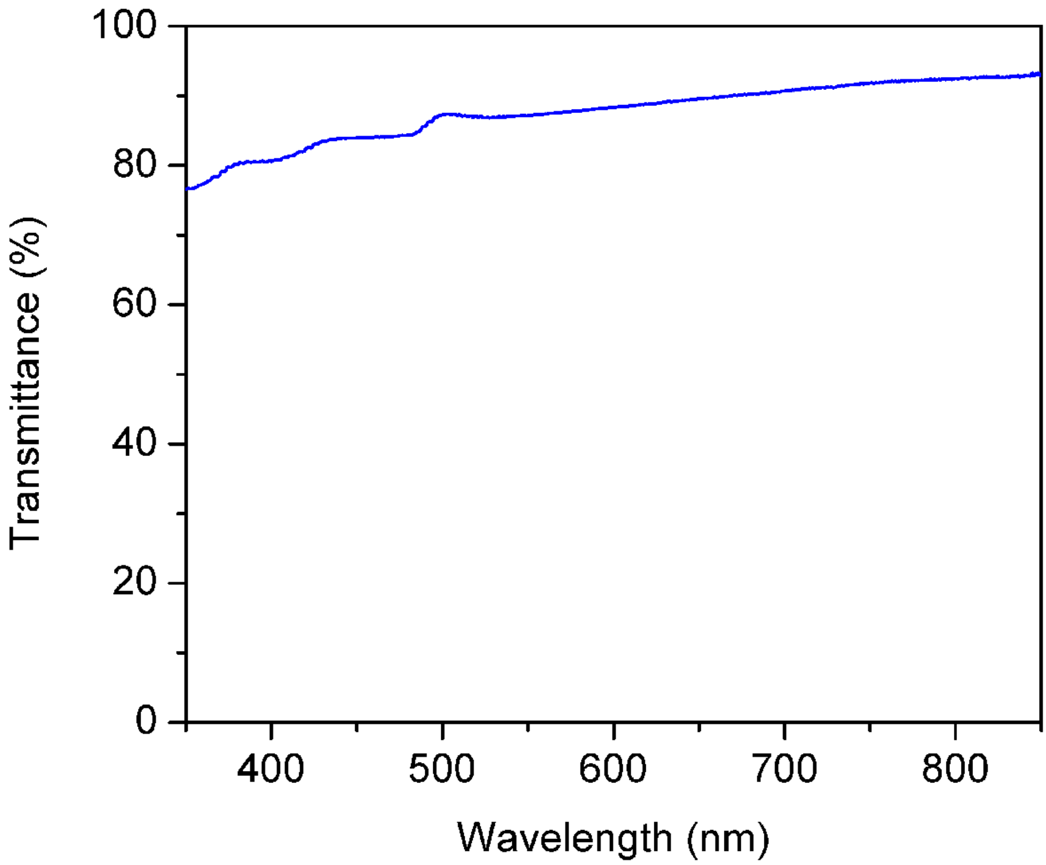

Fig. S3 Dependence of optical transmittance on wavelength for a CNT sheet (thickness of 18 nm).

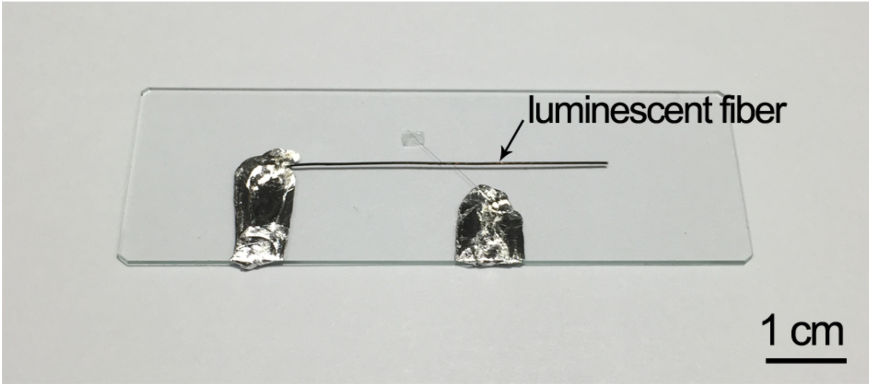

Fig. S4 Photograph of a “luminescent fiber”.

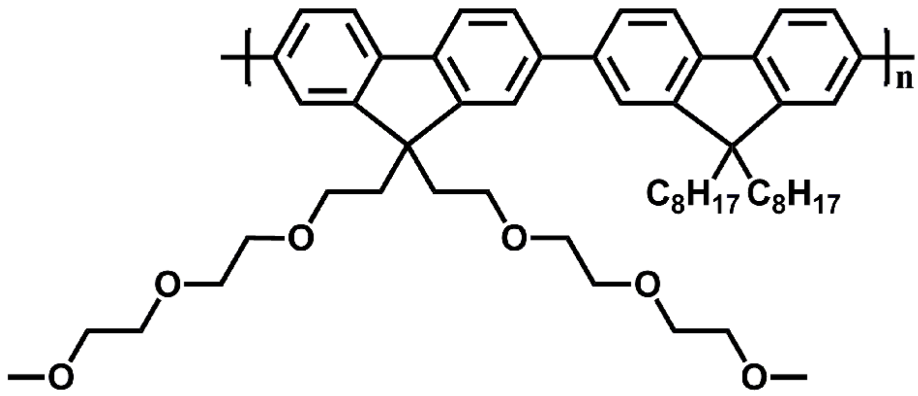

Fig. S5 Chemical structure of the light-emitting conjugated polymer.

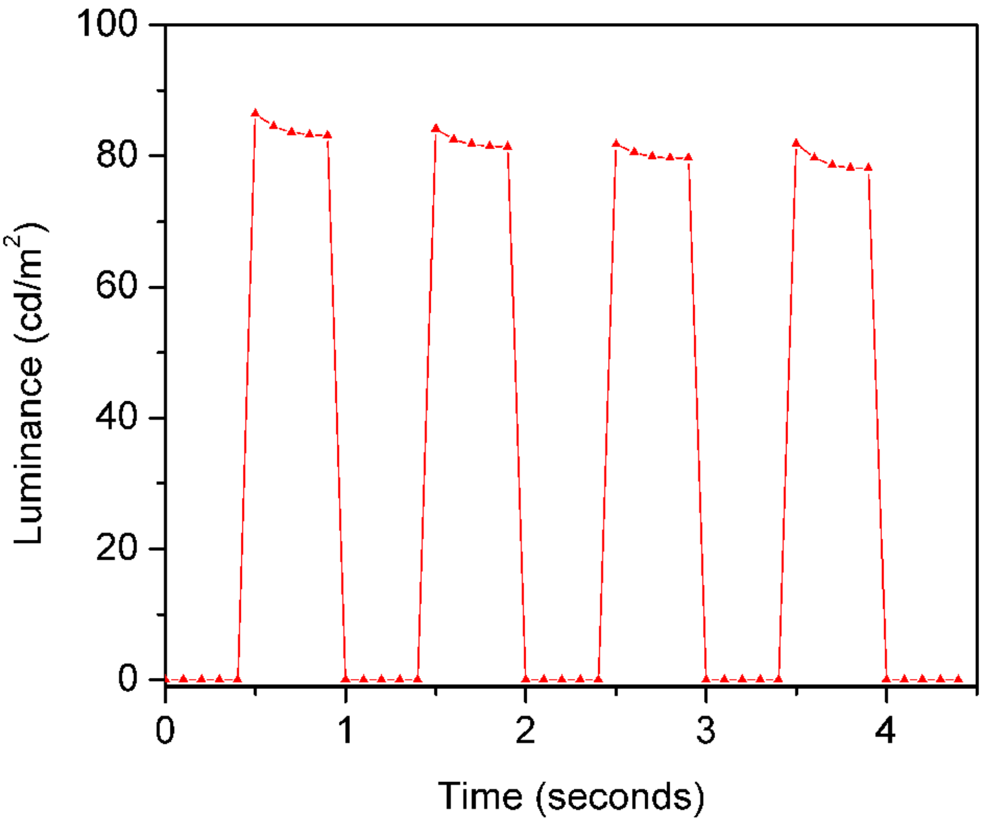

Fig. S6 Transient light emission response under voltage pulse between 0 and 21.5 V with 50% duty cycle for an initially charged "luminescent fiber" (50% duty cycle at 1 Hz).

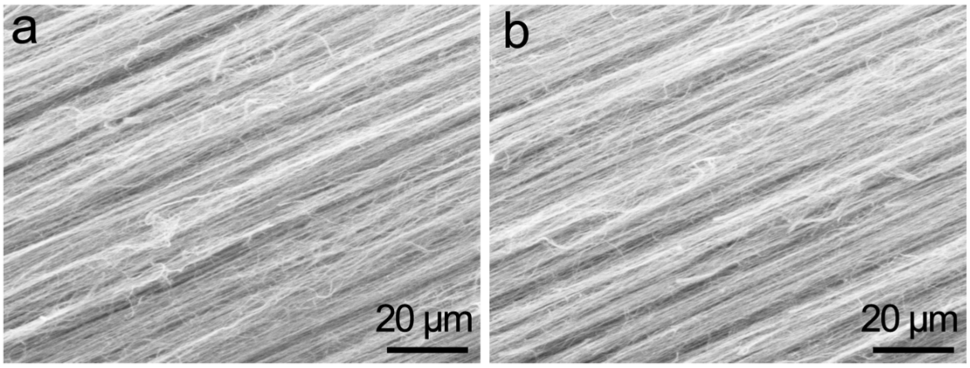

Fig. S7 (a) and (b) SEM images of the outer aligned CNT sheet before and after bending, respectively.

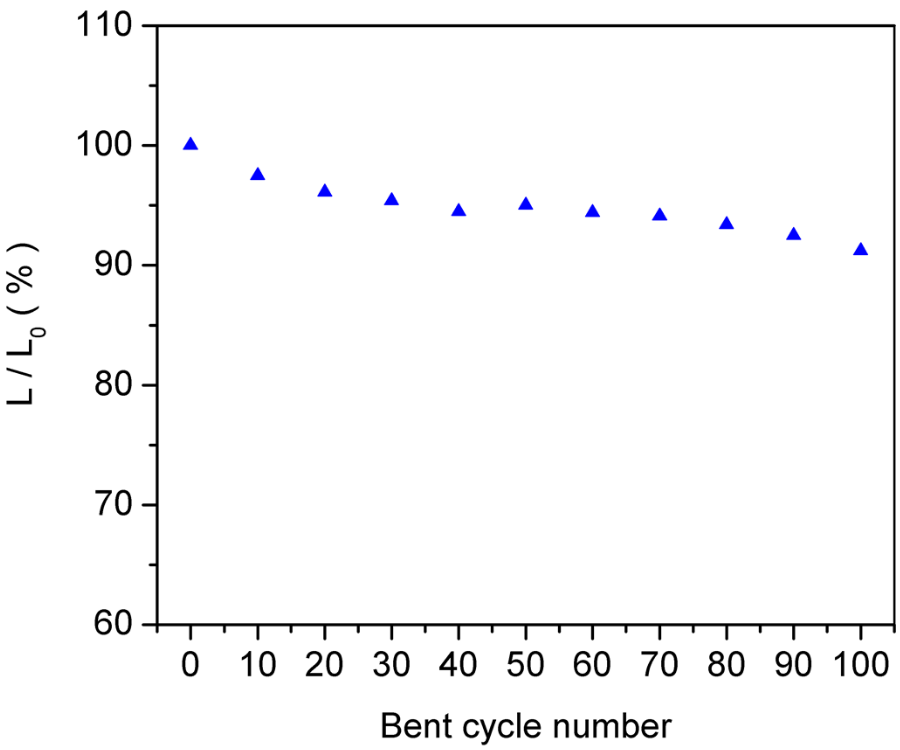

Fig. S8 Dependence of luminance on bending cycle number. Here L_0 and L correspond to the luminance before and after bending, respectively.

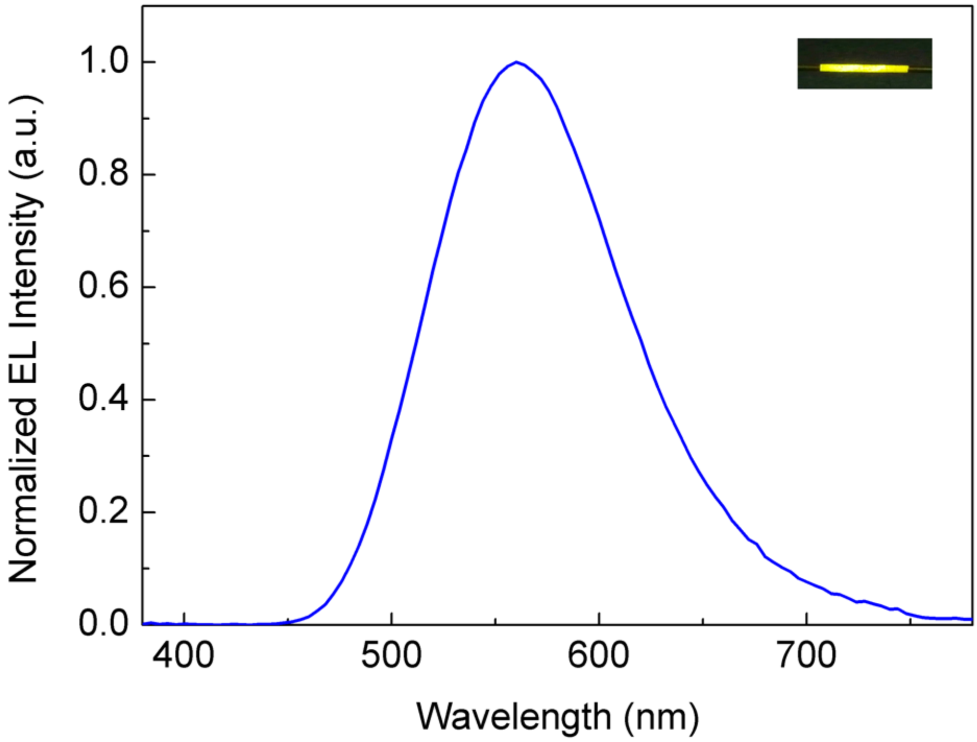

Fig. S9 Polarized electroluminescence spectra of an electroluminescent polymer layer containing SuperYellow, PEO, ETPTA and LiTf (weight ratios of 20/2/2/1). The inserted photograph shows a "luminescent fiber" biased at 15 V.

References for the Supporting Information

[S1] H. Peng, X. Sun, F. Cai, X. Chen, Y. Zhu, G. Liao, D. Chen, Q. Li, Y. Lu, Y. Zhu and Q. Jia, *Nat. Nanotechnol.*, 2009, **4**, 738.

[S2] Z. Zhang, K. Guo, Y. Li, X. Li, G. Guan, H. Li, Y. Luo, F. Zhao, B. Wei, Q. Pei and H. Peng, *Nat. Photon.,* 2015, **9**, 233.