## **Supporting Information**

## **Experimental section**

*Materials.* Poly (3-hexyl thiophene) (P3HT, 99%), phenyl-C<sub>61</sub>-butyric acid methyl ester (PCBM, 99%), tetrahydrofunan (anhydrous, 99.9%, inhibitor-free), zinc acetate dihydrate (99.99%), trimethylolpropane ethoxylate triacrylate (ETT-15) and lithium trifluoromethanesulfonate (LiTf, 99.995%) were obtained from Sigma-Aldrich. The soluble PF-B (polyfluorene copolymer) was obtained from 1-Materials. Methanol (99.9%) and n-butanol (99.8%) was obtained from Aladdin. Potassium hydroxide (85%) and chloroform (99%) were ordered from Sinopharm Chemical Reagent Co., Ltd.

Synthesis of spinnable carbon nanotube (CNT) arrays. Spinnable CNT arrays were synthesized by chemical vapor deposition with Fe/Al<sub>2</sub>O<sub>3</sub> as the catalyst at 740 °C. To prepare the Fe/Al<sub>2</sub>O<sub>3</sub> catalyst, Al<sub>2</sub>O<sub>3</sub> (3 nm) and Fe (1.2 nm) were successively deposited on silicon substrate by electron beam evaporation with rates of 2 and 0.5 Å/s, respectively. Ethylene (C<sub>2</sub>H<sub>4</sub>) and a gas mixture of H<sub>2</sub> and Ar were used as the carbon source and the carrier gas, respectively. The gas flow rates of Ar, C<sub>2</sub>H<sub>4</sub> and H<sub>2</sub> were 400, 90, and 30 cm<sup>3</sup>/min, respectively.

*Characterization.* The structures were characterized by scanning electron microscopy (Hitachi FE-SEM S-4800 operated at 1 kV) and transmission electron microscopy (JEOL JEM-2100F operated at 200 KV). Optical transmittances were recorded by a Shimadzu UV-2550 spectrophotometer. The power conversion efficiencies were measured by recording current density versus voltage characteristic curves with a Keithley 2400 Source Meter under illumination (100 mW/cm<sup>2</sup>) of simulated AM 1.5 solar light coming from a solar simulator (Oriel-Sol3A 94023 A equipped with a 450W Xe lamp and an AM1.5 filter). The current-voltage-light intensity curves were obtained by a Keithley 2400 source meter and a Photoresearch PR-680 by increasing the applied voltage from 5 to 27 V in a 1 V incremental step. The light emission turn-on response curves were measured by a Keithley 2400 source meter and a 21 V incremental step. The light emission turn-on response curves were measured by a Keithley 2400 source meter and a 13 V. All light-emitting measurements were carried out at an argon atmosphere in the glovebox.



Fig. S1. Transmission electron microscopy images of ZnO nanoparticles. a. Low magnification. b. High magnification.



Fig. S2. Chemical structure of the light-emitting conjugated polymer (PF-B).



Fig. S3. Dependence of the sheet resistance of the aligned CNT layer on the thickness.



Fig. S4. Dependence of power conversion efficiency  $(\eta)$  on the thickness of the aligned CNT layer from 18 to 36, 90, 180 and 360 nm.



Fig. S5. Dependence of power conversion efficiency ( $\eta$ ) of the PC part on the time in dried air.



**Fig. S6.** Dependence of the optical transmittance of aligned CNT layer on the thickness (wavelength of 550 nm).



Fig. S7. Dependence of luminance (L) on the thickness of aligned CNT layer.



Fig. S8. Electroluminescence (EL) spectrum of the LE part.



**Fig. S9.** Photoluminescence (PL) spectrum of the light-emitting polymer layer consisted of PF-B, ETT-15 and LiTf (weight ratios of 20/10/1).



**Fig. S10.** Time response of the LE part under voltage pulse between 0 and 13 V (50% duty cycle at 0.25 Hz).