Supplementary Information

Experimental Section

Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) aqueous solution (CLEVIOS P VP Al 4083) was purchased from Bayer Baytron. Hydrothermal titanium dioxide dispersed in terpinol (DHS-TPP3 and DHS-TPP200) was obtained from Dalian Heptachroma Solartech Co., Ltd.

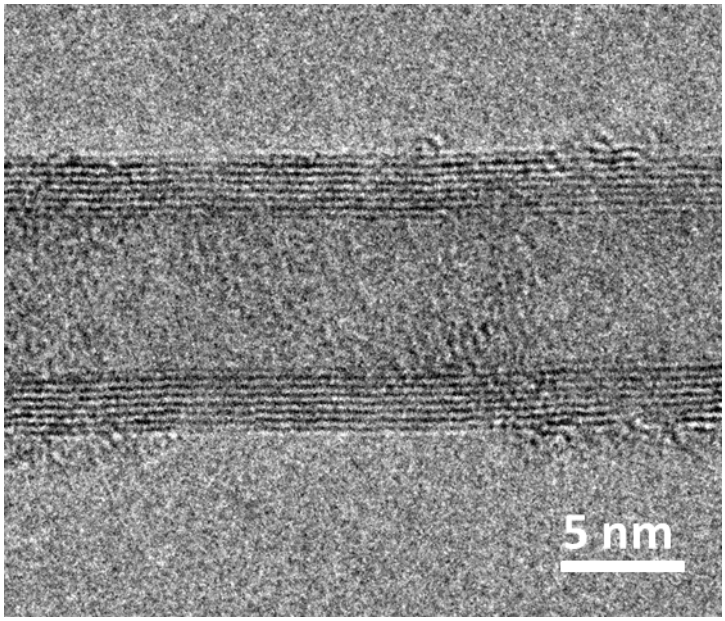

Figure S1. High resolution transmission electron microscopy image of a CNT.

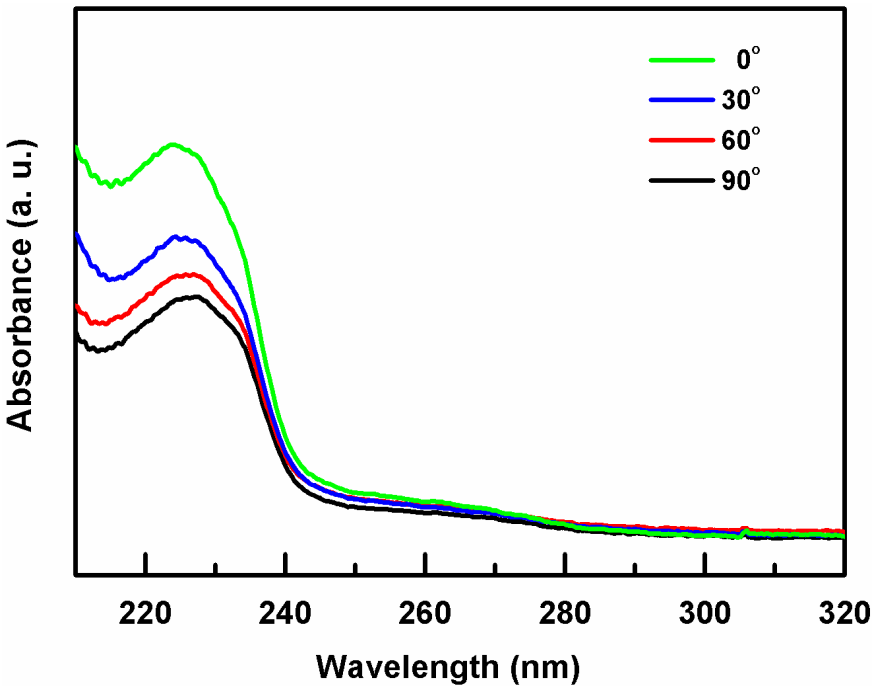

Figure S2. Polarized ultraviolet spectra of the oriented PEDOT:PSS film (250-300 nm in thickness) by a doctor-blading process with included angles of 0°, 30°, 60°, and 90°.

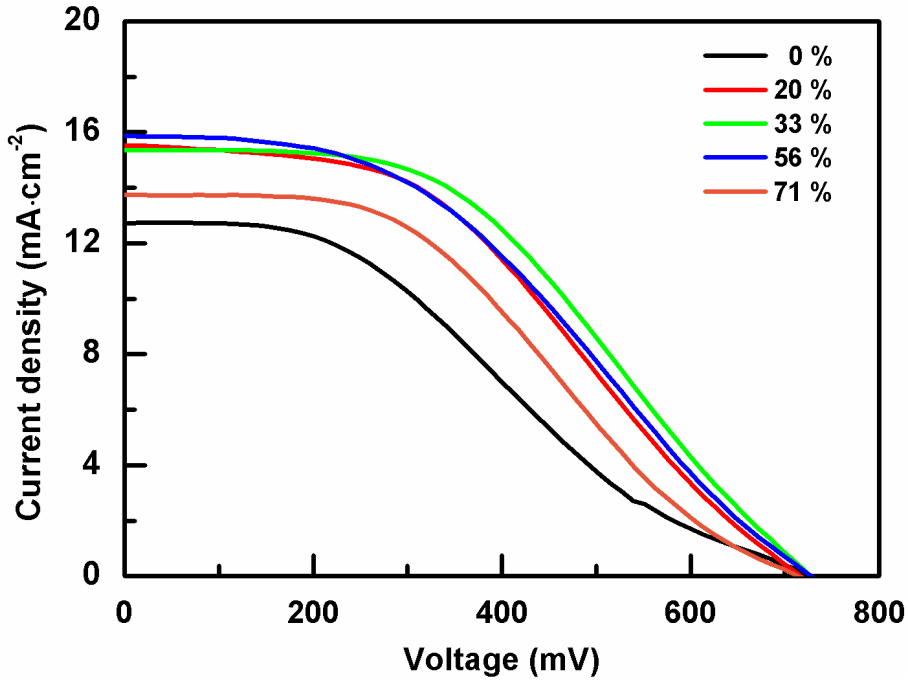

Figure S3. J-V curves of DSCs with increasing polymer weight percentages measured under AM 1.5 illumination. The composite counter electrodes were made by coating the PEDOT:PSS solution on the aligned CNT sheet with thickness of 40 nm.

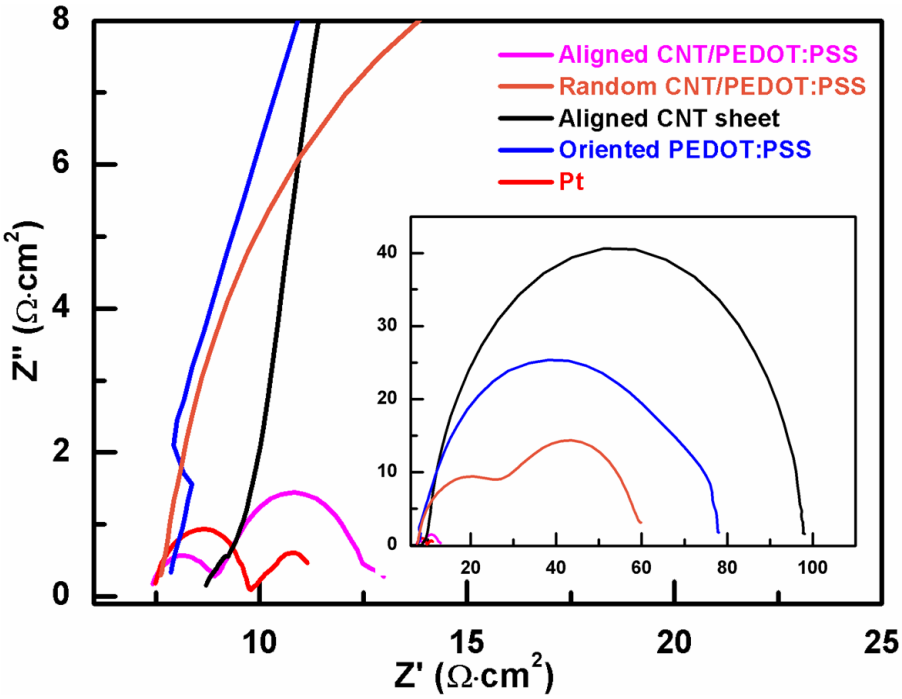

Figure S4. Nyquist plots of different dummy cells that use platinum, aligned CNT sheet, oriented PEDOT:PSS, randomly dispersed CNT/PEDOT:PSS, and aligned CNT/PEDOT:PSS composite with the polymer weight percentage of 33% as counter electrodes.

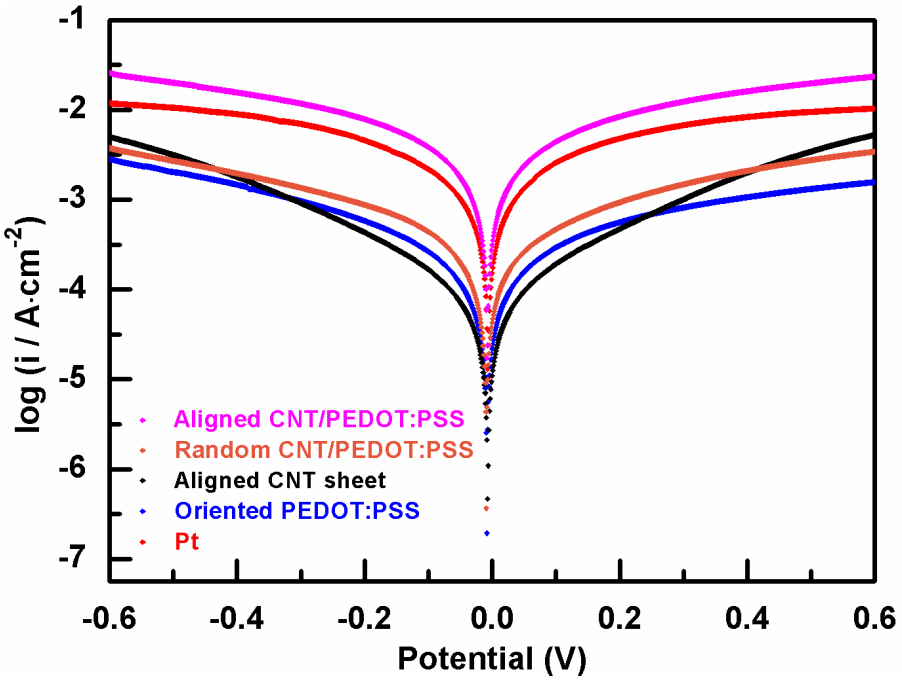

Figure S5. Tafel curves of different dummy cells that use platinum, aligned CNT sheet, oriented PEDOT:PSS, randomly dispersed CNT/PEDOT:PSS, and aligned CNT/PEDOT:PSS composite with the polymer weight percentage of 33% as counter electrodes.

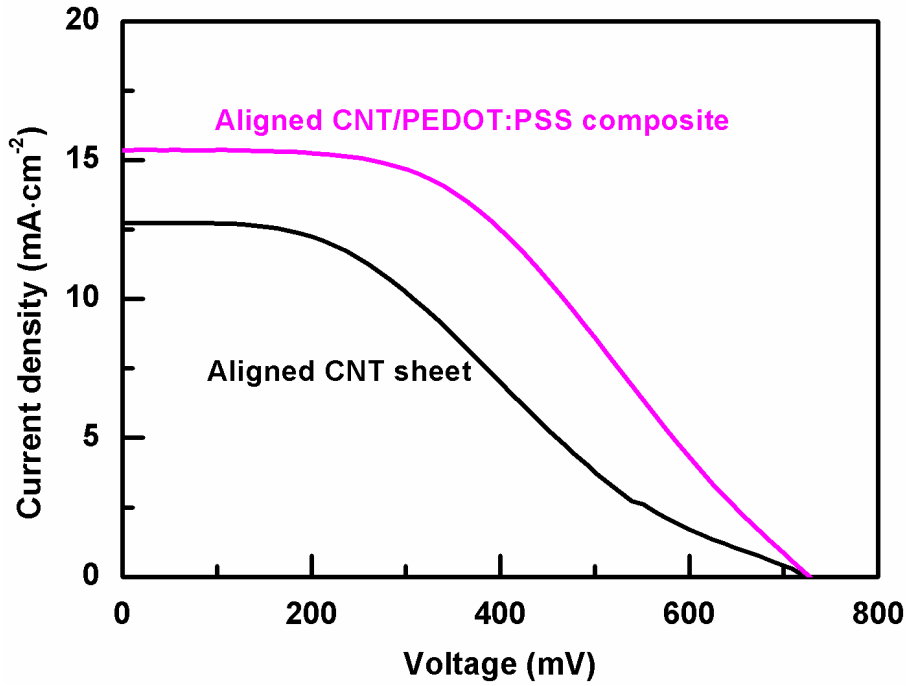

Figure S6. J-V curves for the DSCs before and after coat of PEDOT:PSS onto an aligned CNT sheet with thickness of 40 nm.

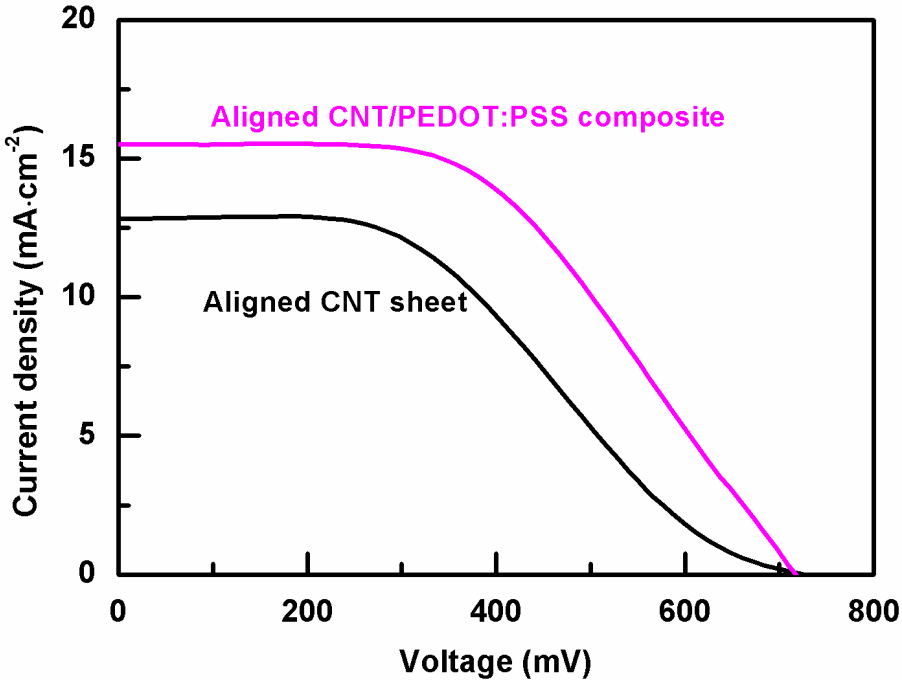

Figure S7. J-V curves for the DSCs before and after coat of PEDOT:PSS onto an aligned CNT sheet with thickness of 200 nm.

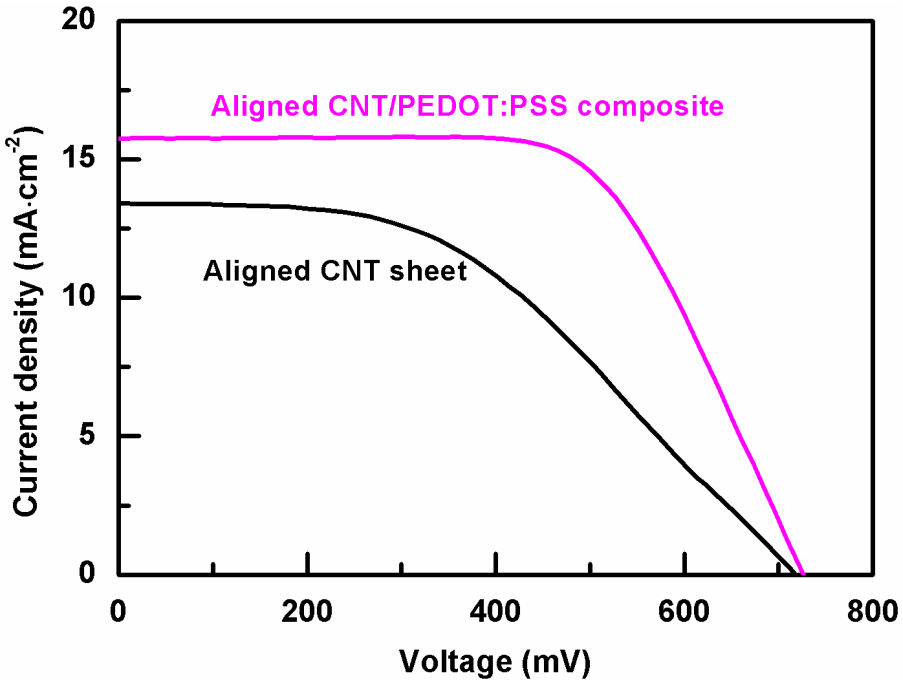

Figure S8. J-V curves for the DSCs before and after coat of PEDOT:PSS onto an aligned CNT sheet with thickness of 1 μm.

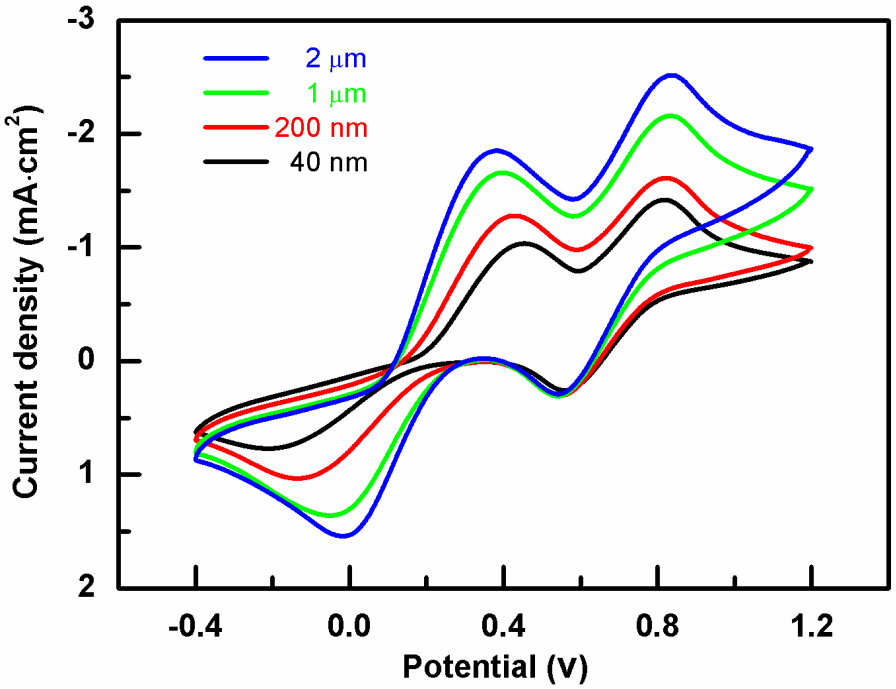

Figure S9. Cyclic voltammograms of aligned CNT sheet/PEDOT:PSS composites with different thicknesses.

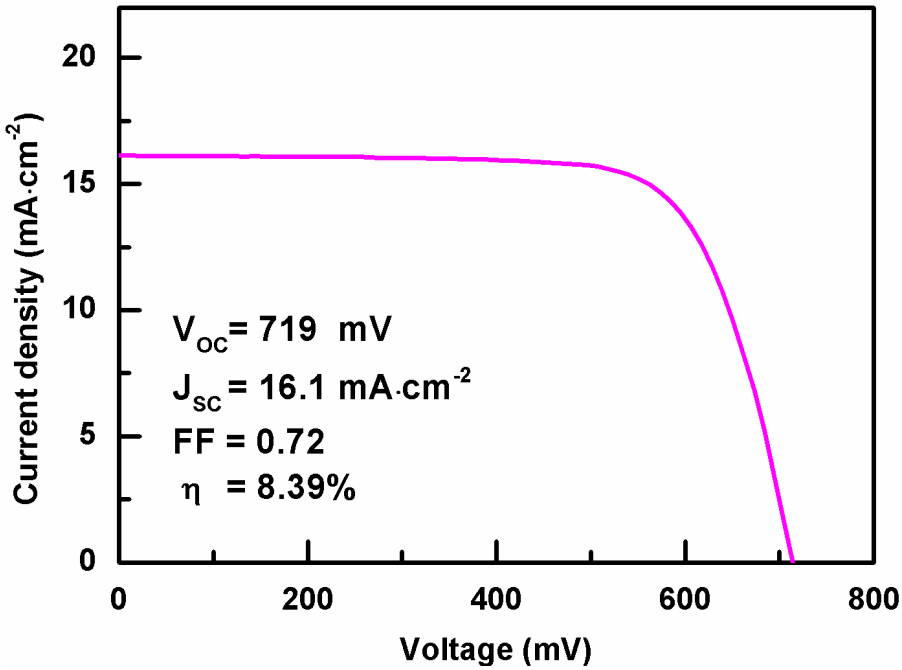

Figure S10. J-V curve of the DSC with the aligned CNT/PEDOT:PSS composite film with a thickness of 4 μm measured under AM 1.5 illumination.

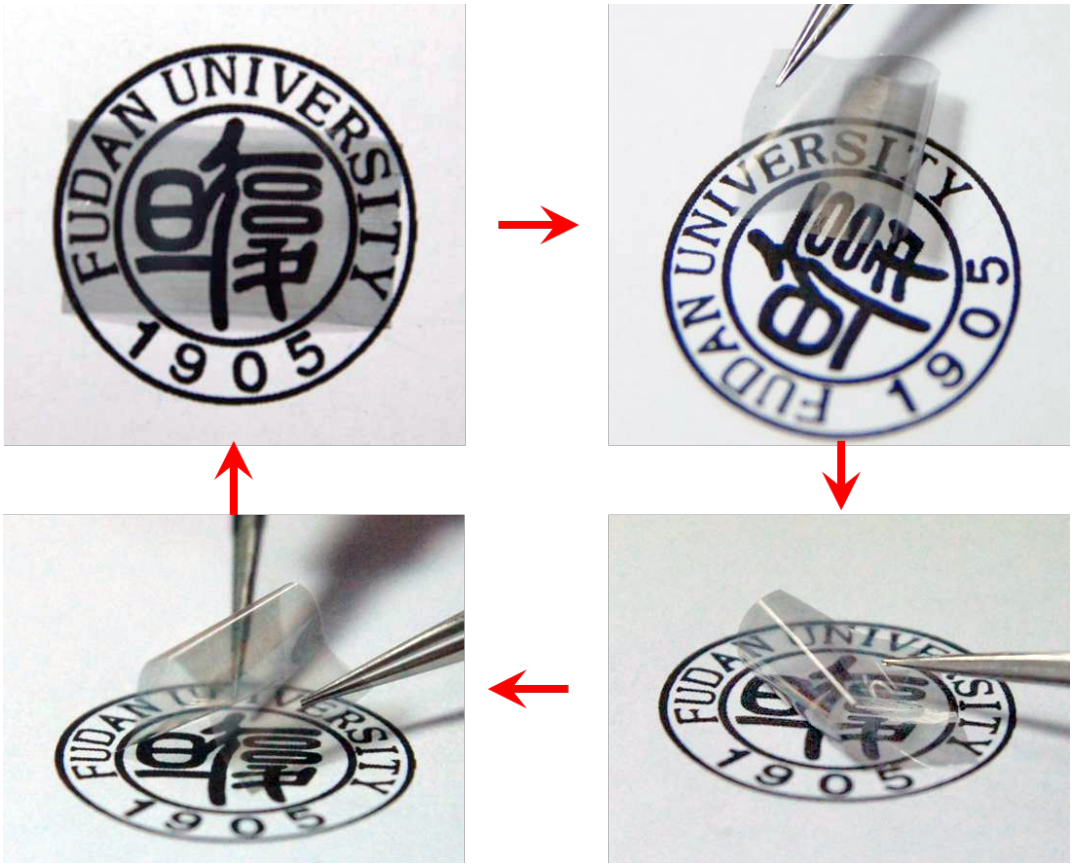

Figure S11. Photographs of a flexible, transparent CNT/PEDOT:PSS composite film with a thickness of 20 nm on polyethylene terephthalate substrate (thickness of 250 μm).

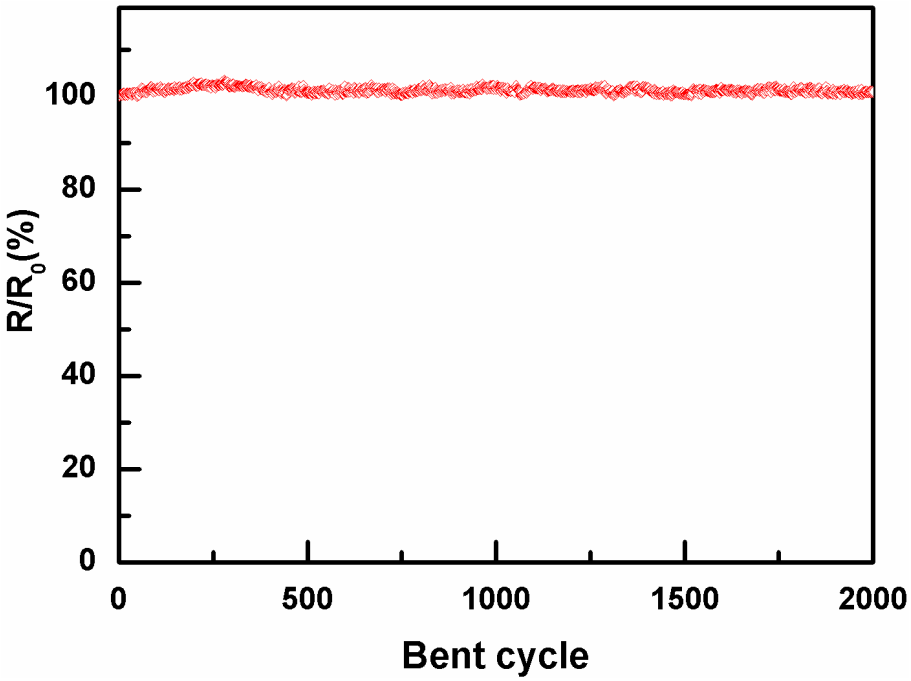

Figure S12. Electrical resistances of a CNT/PEDOT:PSS composite film along the CNT-aligned direction during the deforming process as a function of bent cycle. Here R_0 and R correspond to the electrical resistances before and after bending, respectively.

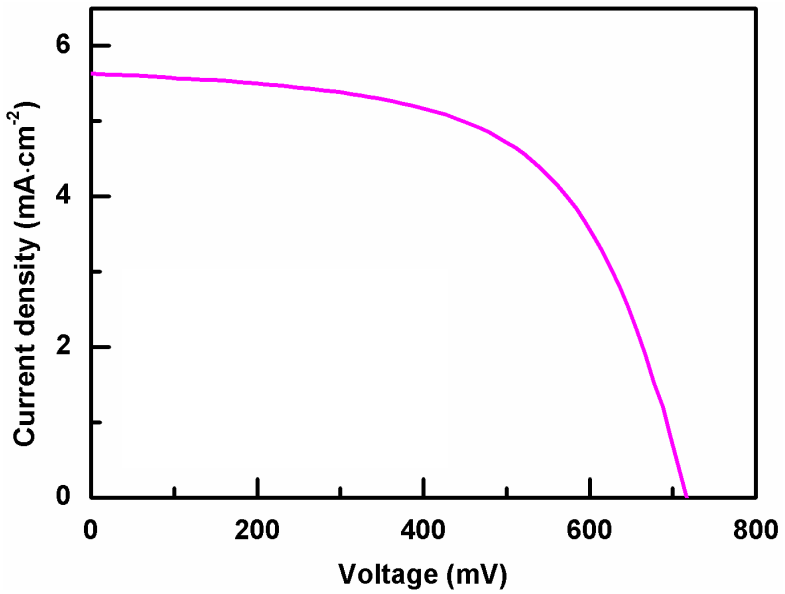

Figure S13. J-V curves of a flexible DSC using the aligned CNT/PEDOT:PSS composite as the counter electrode measured under AM 1.5 illumination. The composite counter electrode was made by coating the PEDOT:PSS solution on the aligned CNT sheet with thickness of 2 μm.

Table S1. Photovoltaic parameters of DSCs using aligned CNT/PEDOT:PSS composites by coating PEDOT:PSS solutions on the same aligned CNT sheet with thickness of 40 nm with increasing polymer weight percentages in Figure S3.

Polymer content (%)	V_{OC} (mV)	J_{SC} (mA/cm^2)	FF	η (%)
0	0.719	12.7	0.34	3.3
20	0.720	15.5	0.40	4.6
33	0.720	15.3	0.45	5.1
56	0.723	15.8	0.39	4.7
71	0.718	13.7	0.41	3.9

Table S2. EIS parameters of the dummy cells shown in Figures S4 and S5.

Counter electrode	R_s ($\Omega \bullet cm^2$)[a]	R_{ct} ($\Omega \bullet cm^2$)[b]	J_0 (mA/cm^2)	J_0 [c] (mA/cm^2)
Aligned CNT/PEDOT:PSS	7.4	1.63	7.8	5.4
Random CNT/PEDOT:PSS	7.7	10.8	1.2	0.68
Oriented PEDOT:PSS	7.9	35.3	0.36	0.21
Aligned CNT sheet	8.7	44.7	0.29	0.15
Pt	7.5	2.56	5.0	3.9

[a] R_s=series resistance. [b] R_{ct}=charge transfer resistance.

[c] The data were calculated from the Tafel curves in Figure S5.

Table S3. Photovoltaic parameters of the DSCs for the counter electrodes before and after coat of the same PEDOT:PSS on the aligned CNT sheets with increasing thicknesses (δ) from Figures S6-S8.

Counter electrode	V_{OC} (V)	J_{SC} (mA/cm^2)	FF	η (%)
Sheet (δ=40 nm)	0.719	12.7	0.34	3.3
Sheet (δ=200 nm)	0.726	12.8	0.39	4.0
Sheet (δ=1 μm)	0.721	13.4	0.46	4.5
Sheet (δ=2 μm)	0.733	13.5	0.50	5.0
Composite (δ=40 nm)	0.720	15.3	0.45	5.1
Composite (δ=200 nm)	0.718	15.5	0.51	5.8
Composite (δ=1 μm)	0.723	15.8	0.63	7.4
Composite (δ=2 μm)	0.731	16.1	0.71	8.3