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Supporting Information

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Carbon Nanotubes Bridged with Graphene Nanoribbons and Their Use in High-Efficiency Dye-Sensitized Solar Cells**

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Supporting Information

Experimental Section

Materials. Multi-walled CNTs with wall numbers of 20-30 were supplied by Chengdu Institute of Organic Chemistry, Chinese Academy of Sciences, China. Hydriodic acid (HI) with a concentration of 55% was purchased from Sigma-Aldrich. Fluorine-doped tin oxide conductive glass was obtained from Nippon Sheet Glass Co., Japan. Poly (ethylene naphthalate) was obtained from Peccell Co., Japan. Titanium dioxide slurry (DHS-TPP3 and DHS-TPP200, hydrothermal titanium dioxide dispersed in terpinol) were obtained from Dalian Heptachroma Solartech Co., Ltd. All other chemicals were ordered from Sinopharm Chemical Reagent Co. Ltd and used as received.

Preparation of acid-treated CNTs. They were prepared by refluxing the pristine multi-walled CNTs in a concentrated nitric acid for 5 h at 100 ^{o}C. The mixture was filtered and washed by 5% hydrochloric acid after cooling down to room temperature. The resulting solid was then dialyzed against deionized water until the pH was close to 7. The resulting solution was dried in a vacuum oven at 70 ^{o}C for 24 h to obtain the acid-treated CNTs.

Characterization. Transmission electron microscopy (TEM) was performed on a Jeol JEM 2100 with an accelerating voltage of 200 kV. The samples were prepared by drop-casting samples onto 300 mesh carbon grids on a copper support (Ted Pella). Scanning electron microscopy (SEM) was performed at a Hitachi FE-SEM S-4800 operated at 1 kV. X-ray diffraction measurements (XRD) were made by a PANalytical X'Pert PRO with Cu Kα radiation (λ = 0.1542 nm; operating energy, 40 kV; cathode current, 40 mA; scan rate, 2^{o} min^{-1}). Fourier transform infrared (FTIR) spectra were obtained from a Nicolet Nexus 470 infrared spectrophotometer using KBr discs with a scan range of 400-4000 cm^{-1} and signal-averaging 64 scans at a resolution of 4 cm^{-1}. Raman spectra were collected on an Avalon Instruments Raman Station using a 632.8 nm He-Ne laser. The XPS spectra were collected by a Perkin Elmer PHI 5000 C ESCA spectrometer equipped with a hemispherical electron energy analyzer at a pressure lower than 1029 Torr. The Mg-Ka (hu = 1253.6 eV) anode was operated at 14 kV and 20 mA. The carbonaceous C 1s line (284.6 eV) was used as the reference to calibrate the binding energies. Thermogravimetric analysis (TGA) was performed from 100 to 800 ℃ with a heating rate of 20 ℃ min^{-1} by using a Perkin Elmer Pyris-1 under nitrogen atmosphere. The film thickness was measured by Dektak 150 Step Profiler. The J-V curves of DSCs were measured by a Keithley 2400 Source Meter under illumination (100 mW/cm^{2}) of simulated AM1.5 solar light

coming from a solar simulator (Oriel-Sol3A 94023A equipped with a 450 W Xe lamp and an AM1.5 filter). The light intensity was calibrated using a reference Si solar cell (Oriel-91150). Cyclic voltammetry were performed on CHI 660a electrochemical workstation. The sheet resistance is measured by four-probe method with a 4-point probes resistivity measurement system (RTS-8).

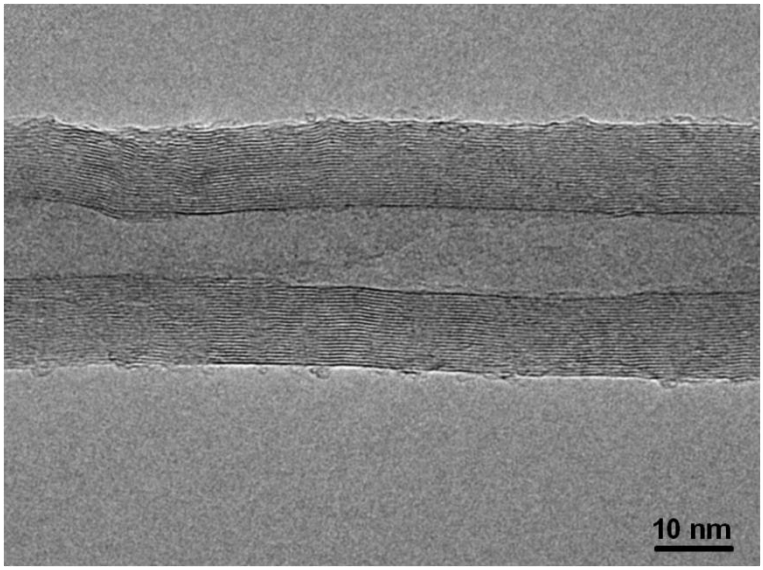

Figure S1. TEM image of a typical CNT.

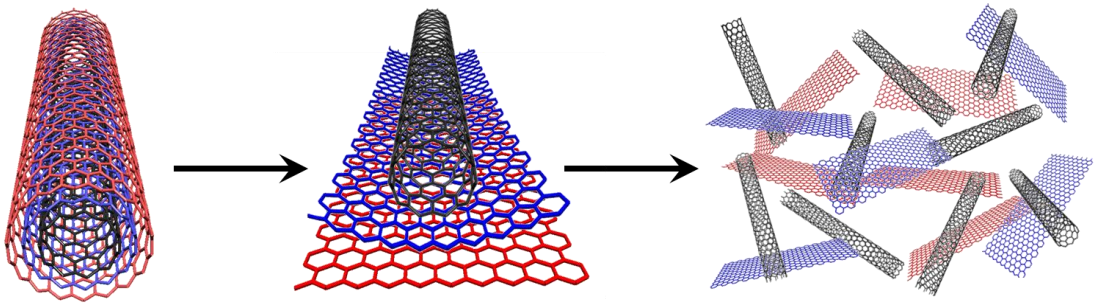

Figure S2. Schematic illustration to the formation of the GONR/CNT hybrid.

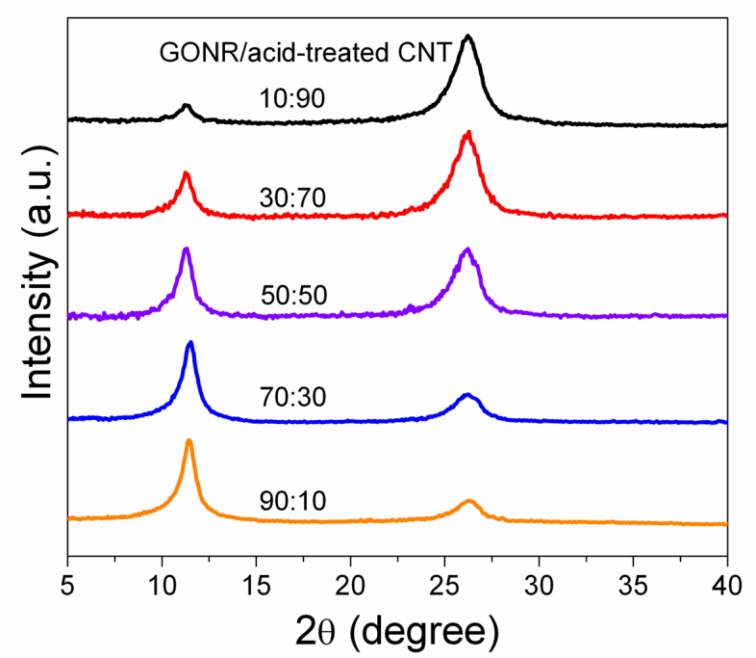

Figure S3. XRD patterns of GONR/acid-treated CNT mixtures with different GONR weight percentages.

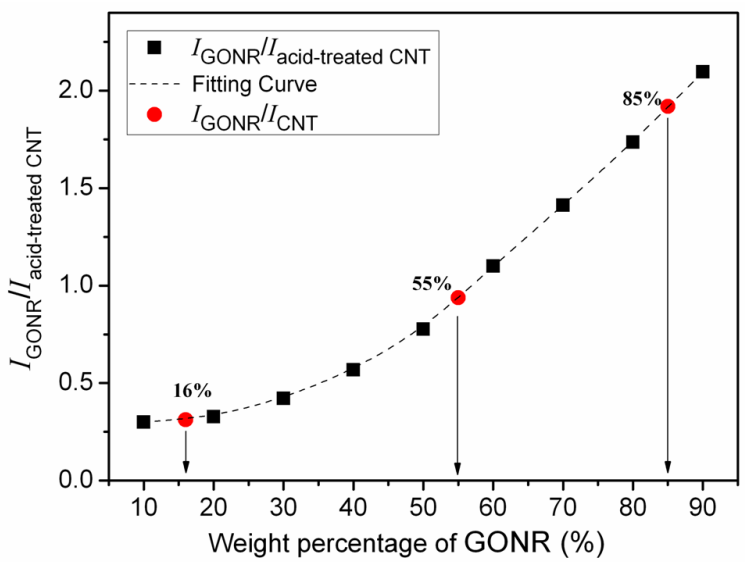

Figure S4. Calculation of the GONR weight percentage in GONR/CNT hybrid materials according to the relationship curve between GONR weight percent and intensity ratio of characteristic peaks in the XRD spectra. $I_{GONR}/I_{acid\text{-}treated\ CNT}$ corresponds to the intensity ratio of characteristic XRD peaks of GONR and acid-treated CNTs in their mixtures, while I_{GONR}/I_{CNT} corresponds to the intensity ratio of GONR and CNTs in the hybrid materials.

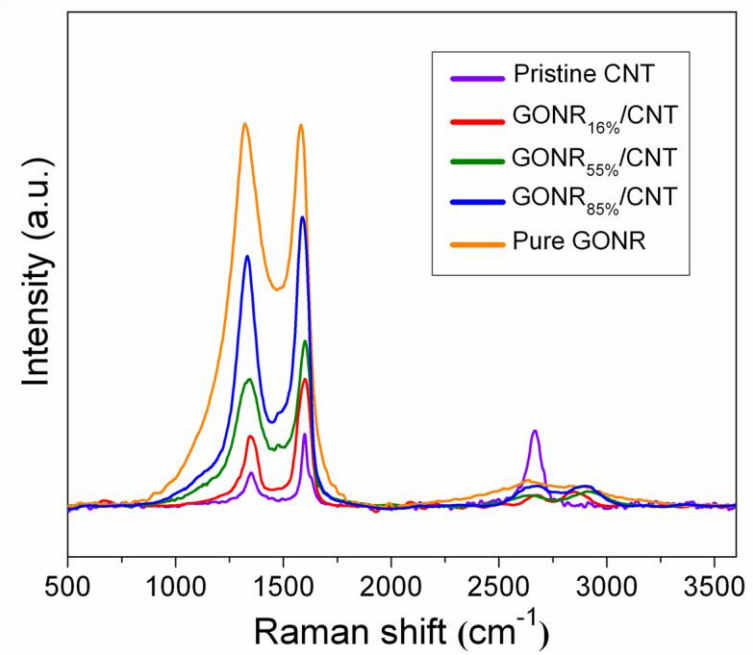

Figure S5. Raman spectra of pristine CNTs, GONR/CNT hybrids with different GONR weight percentages, and pure GONR (excitation at 632.8 nm).

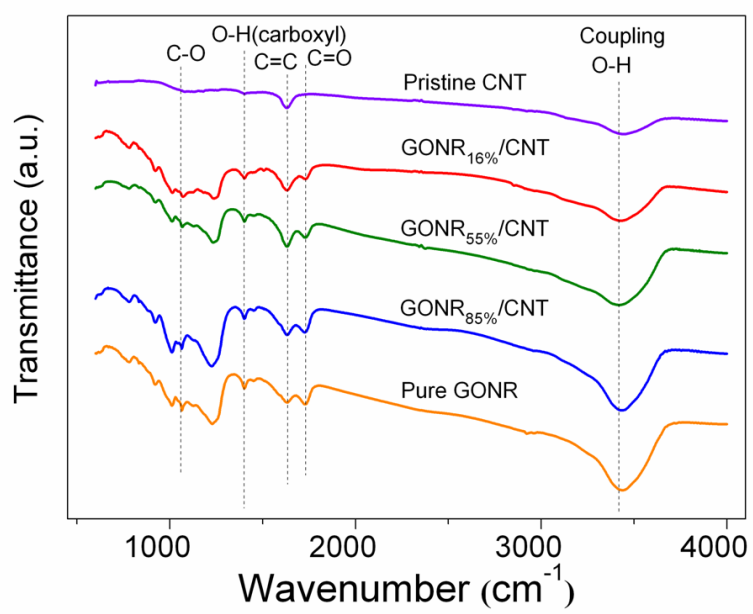

Figure S6. FTIR spectra of pristine CNTs, GONR-CNT hybrids with different GONR weight percentages, and pure GONR.

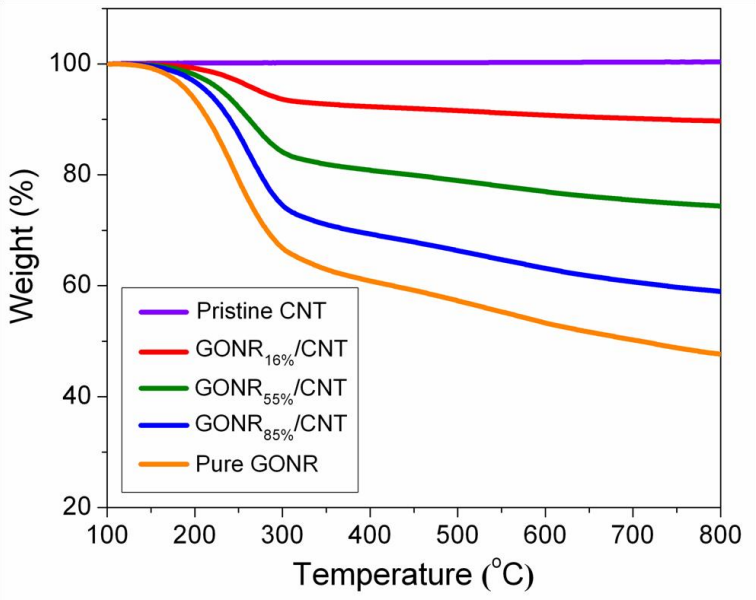

Figure S7. TGA curves of pristine CNTs, GONR/CNT hybrids with different GONR weight percentages, and pure GONR.

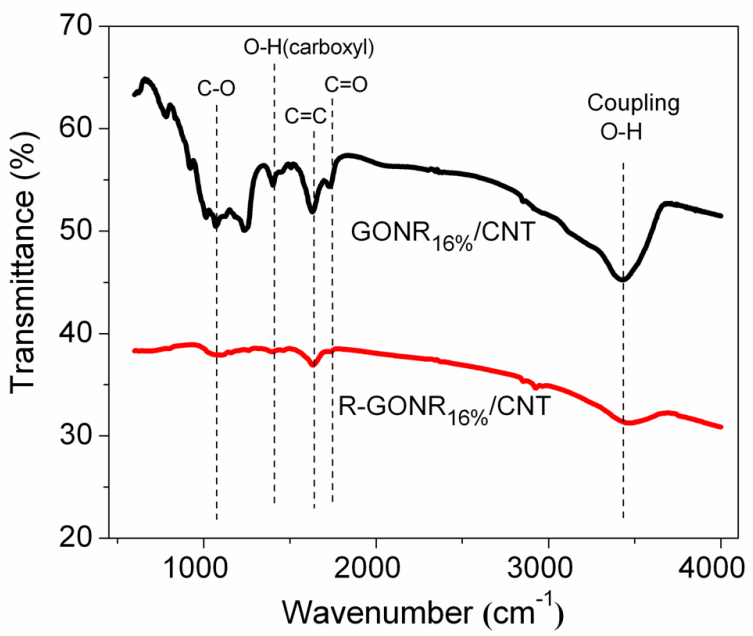

Figure S8. FTIR spectra of $GONR_{16\%}$/CNT and R-$GONR_{16\%}$/CNT hybrids.

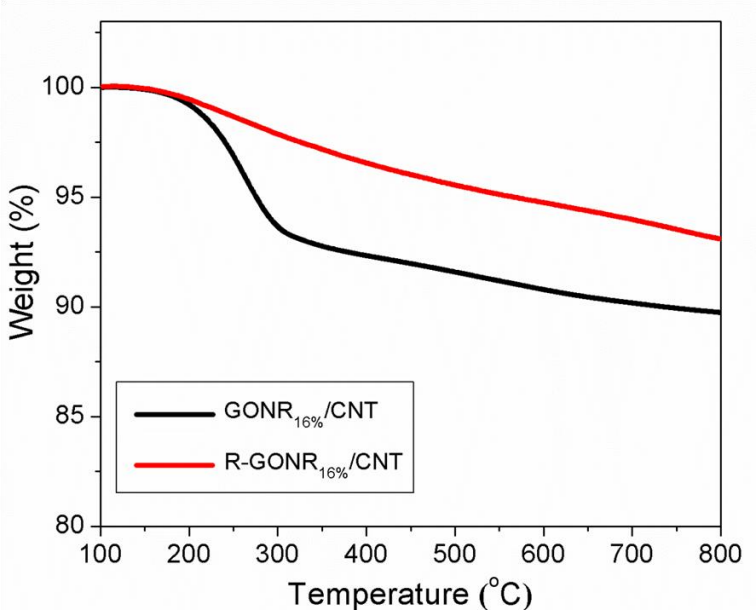

Figure S9. TGA curves of $GONR_{16\%}$/CNT and R-$GONR_{16\%}$/CNT hybrids.

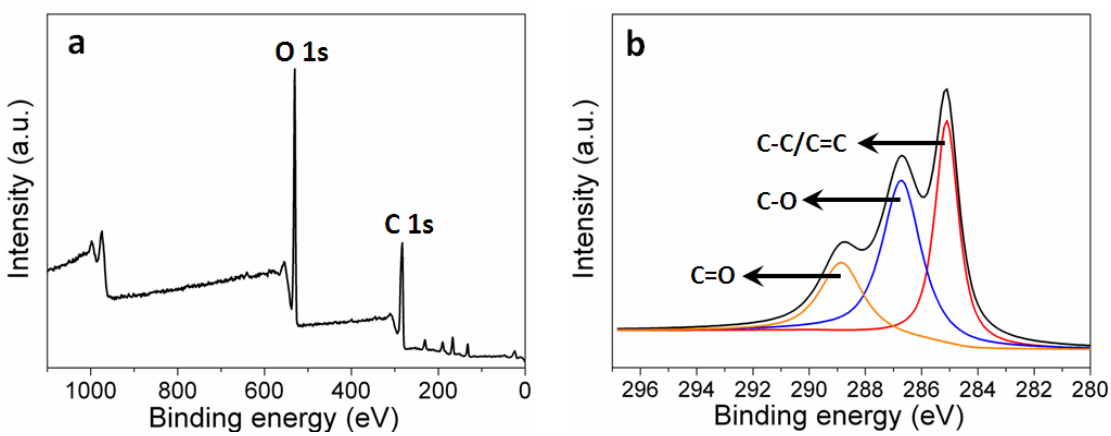

Figure S10. X-ray photoelectron spectroscopy (XPS) characterization of the $GONR_{16\%}$/CNT hybrid material.

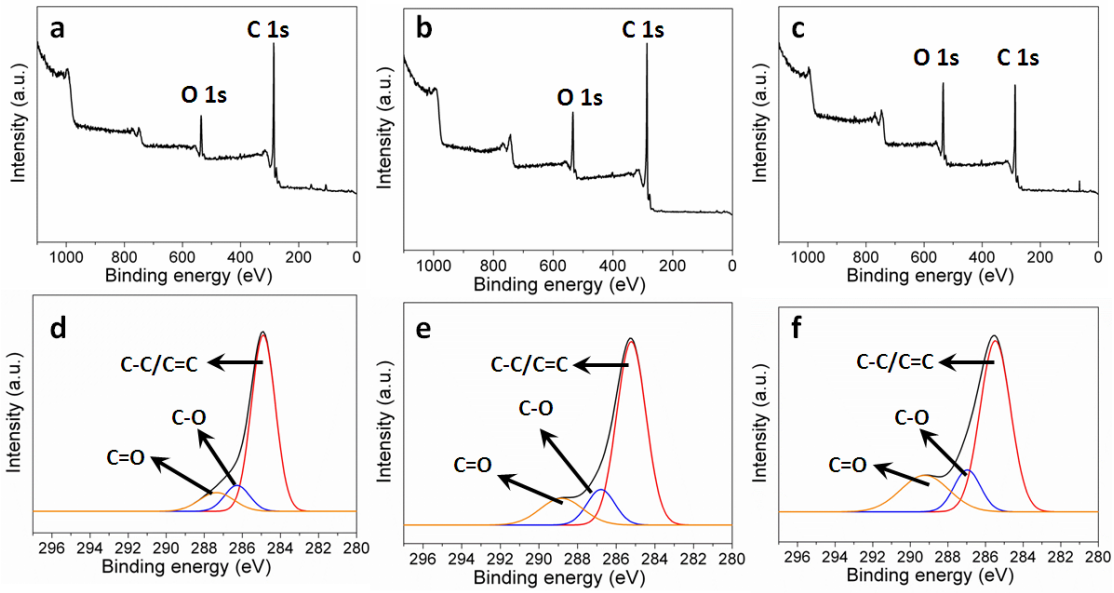

Figure S11. XPS spectra of (a, d) pristine CNTs, (b, e) R-GONR$_{16\%}$/CNT hybrid, and (c, f) pure R-GONRs.

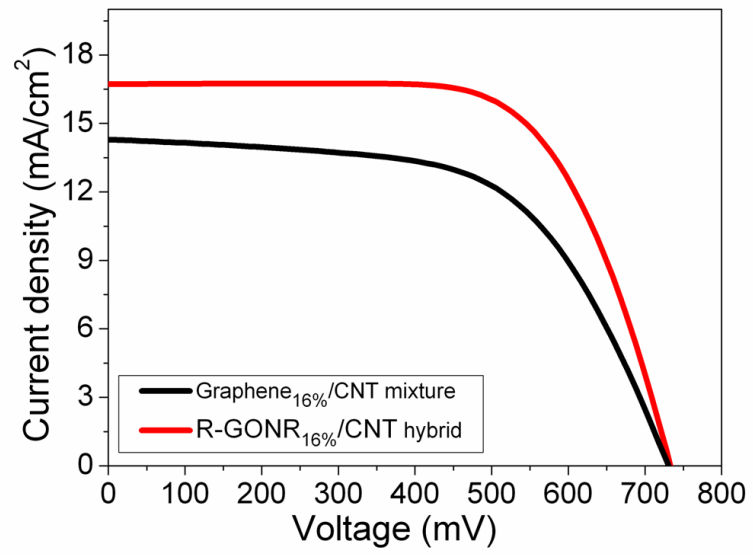

Figure S12. J-V characteristics of the DSC by using the R-GONR/CNT hybrid and graphene/CNT mixture with the same weight percentage of 16% for R-GONR and graphene as counter electrodes measured under AM 1.5 illumination.

Table S1. Parameters of the DSC by using pure CNTs, R-GONR/CNT hybrids with increasing R-GONR weight percentages, pure R-GONR, and platinum as counter electrodes measured under AM 1.5 illumination.

Counter electrode	V_{OC} (mV)	J_{SC} (mA/cm^2)	FF	η (%)
Pure CNTs	0.727	15.23	0.55	6.09
$GNR_{16\%}$/CNT	0.734	16.73	0.67	8.23
$GNR_{55\%}$/CNT	0.729	16.21	0.58	6.85
$GNR_{85\%}$/CNT	0.730	14.33	0.38	3.98
Pure GNR	0.729	14.17	0.32	3.31
Platinum	0.731	16.53	0.63	7.61

Table S2. Parameters of the DSC by using R-GONR$_{16\%}$/CNT hybrids with increasing thicknesses as counter electrodes measured under AM 1.5 illumination.

Thickness of counter electrode	V_{OC} (mV)	J_{SC} (mA/cm^2)	FF	η (%)
50 nm	0.729	12.98	0.27	2.55
100 nm	0.732	13.66	0.38	3.80
200 nm	0.735	15.11	0.49	5.44
300 nm	0.733	15.81	0.58	6.72
500 nm	0.734	16.73	0.67	8.23
1 μm	0.731	16.53	0.68	8.22

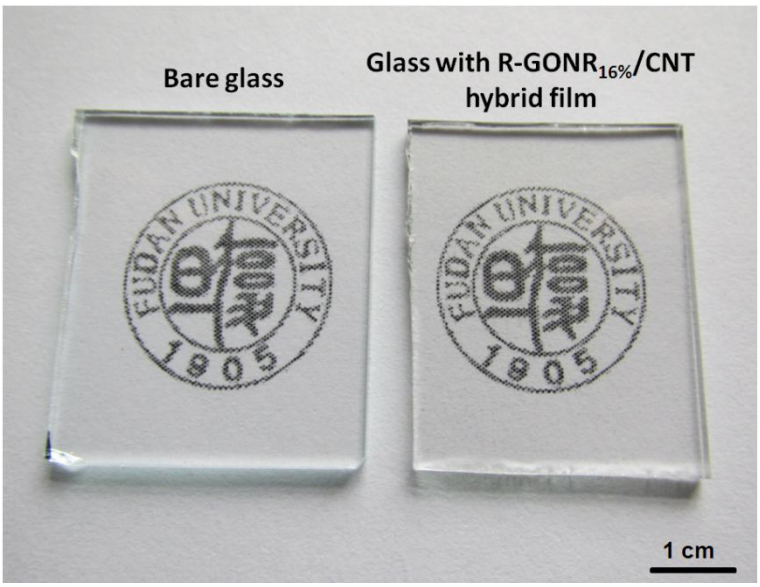

Figure S13. Photographs of only a glass slide and a glass slide with the R-GONR$_{16\%}$/CNT hybrid film on one side on a paper which was marked with the logo of Fudan University. The hybrid film showed a thickness of 50 nm.

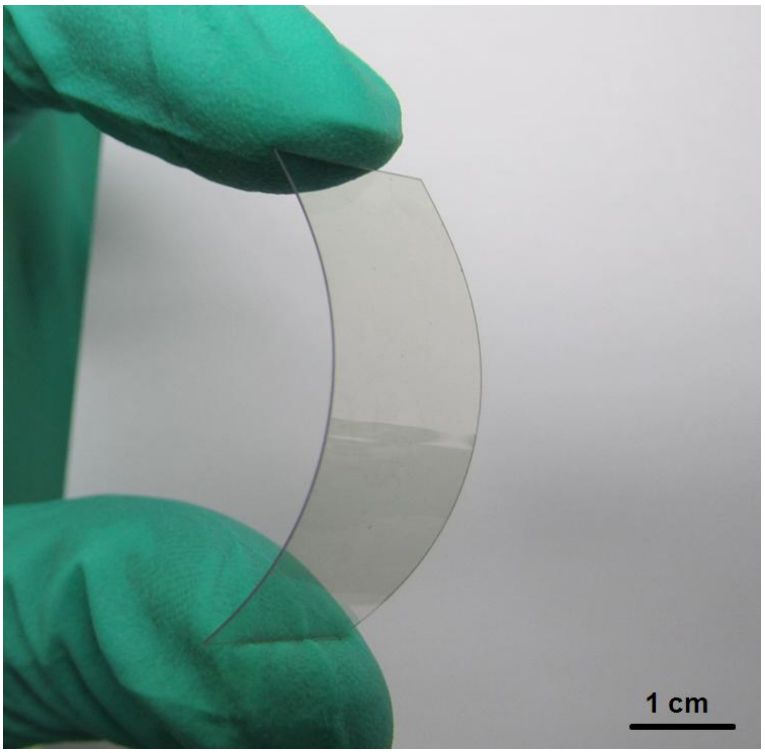

Figure S14. Photograph of a flexible R-GONR$_{16\%}$/CNT hybrid film on the poly (ethylene naphthalate) substrate. The hybrid film showed a thickness of 50 nm.

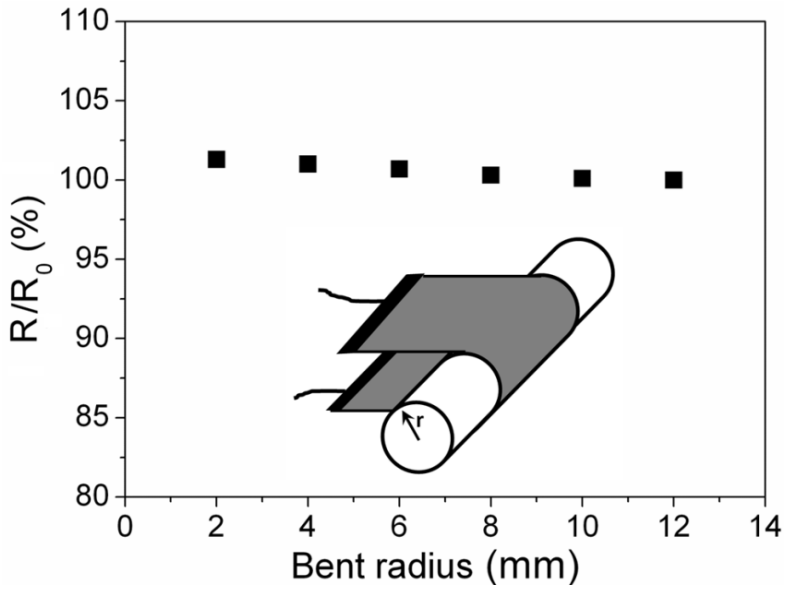

Figure S15. The resistance change of a flexible R-GONR$_{16\%}$/CNT hybrid film on the poly (ethylene naphthalate) substrate during the bending process. Here R_0 and R correspond to the resistances before and after bending, respectively. The inserted image schematically shows the bending process. The hybrid film was deformed with different bending radii in a range from 2 to 12 mm.

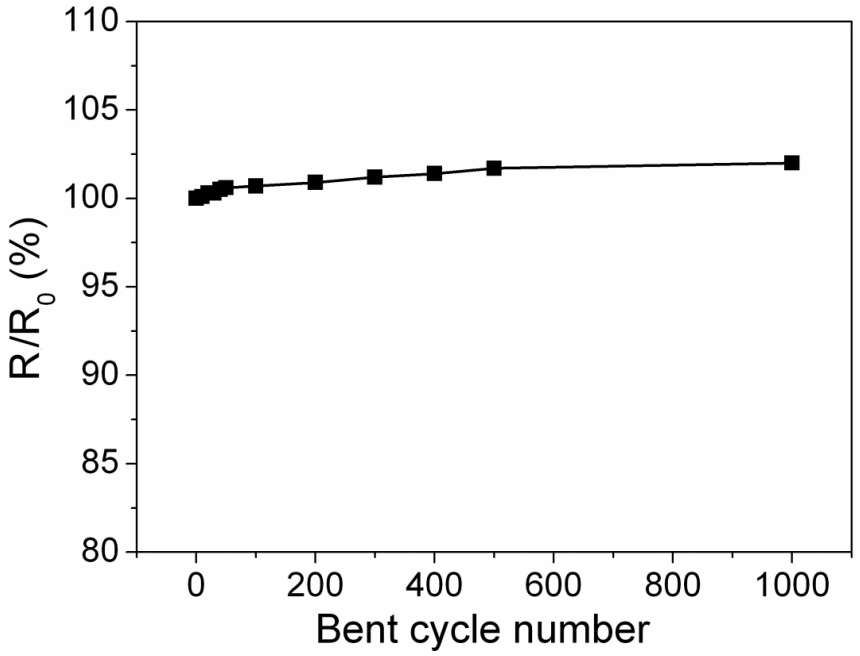

Figure S16. The resistance dependence on the bent cycle number for a flexible R-GONR$_{16\%}$/CNT hybrid film on the poly (ethylene naphthalate) during the bending process. R_0 and R correspond to the resistances before and after bending, respectively. The film resistance was changed in less than 2% even after bending for 1000 cycles.

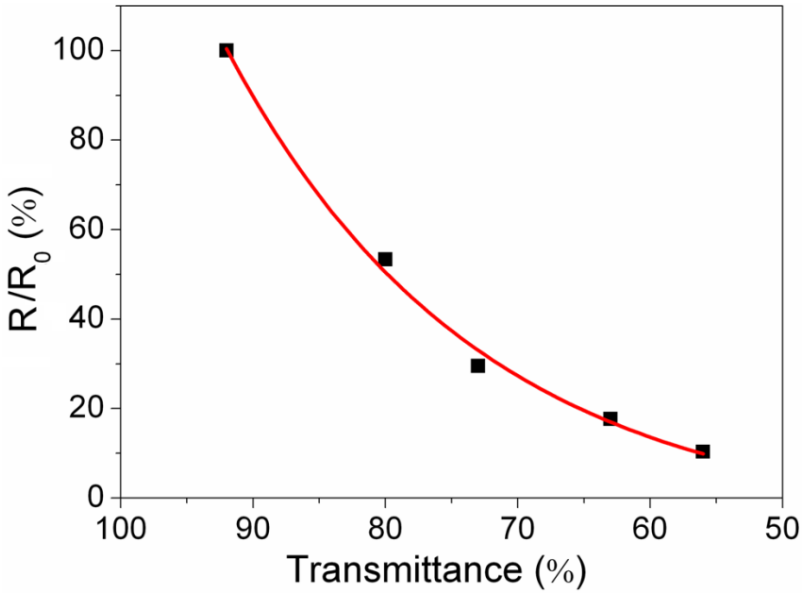

Figure S17. The resistance change with the transmittance in the R-GONR$_{16\%}$/CNT hybrid film. R_0 and R correspond to the resistances at the highest transmittance of 92% and other transmittances, respectively.

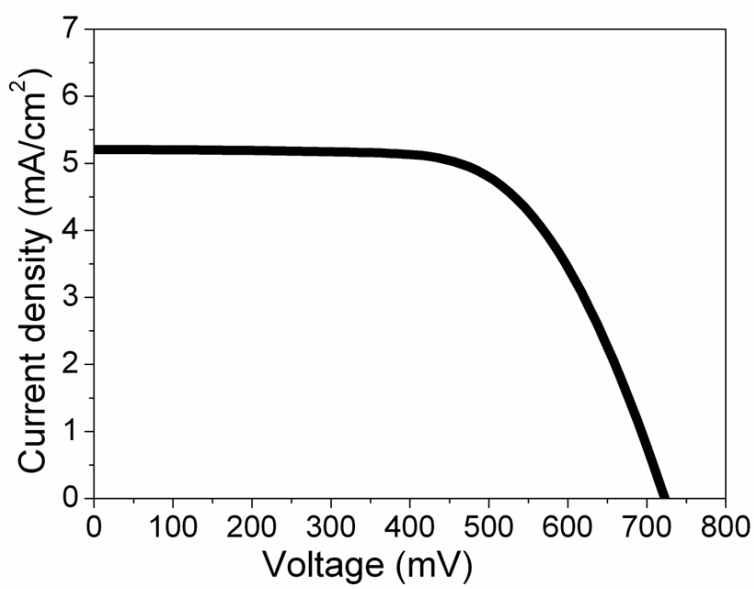

Figure S18. Typical J-V curve of a flexible dye-sensitized solar cell by using a flexible R-GONR$_{16\%}$/CNT hybrid film as the counter electrodes measured under AM 1.5 illumination. The resulting cell exhibited V_{OC} of 0.72V, J_{SC} of 5.20 mA cm^{-2}, and FF of 0.65, which produced a η of 2.44 %.