

Supporting Information

Carbon dots and AIE molecules for highly efficient tandem luminescent solar concentrators

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1. EXPERIMENTAL

1.1 Preparation of N-CDs

N-CDs were synthesized through a simple, convenient and one step method. The preparation was referred to our previous report.¹ Briefly, 3.0 g of citric acid and 2.0 g of urea were added to 10 mL ultrapure water to form a transparent solution under stirring. Then, the solution was transferred to the microwave oven (800 W) and heated for 5 min. The colorless clear reaction solution would become brown solution and finally a clustered

solid. The resulting N-CDs were left to cool, diluted with water and then with a dialysis membrane filter (0.22 μm) in order to eliminate suspended impurities.

1.2 Preparation of TPFE-Rho

TPFE-Rho was prepared following the synthetic procedure reported in the literature.² First, Rhodanic-CN and TPFE-CHO were prepared. Then, 1.21 g of Rhodanic-CN, 1.75 g of TPFE-CHO, 0.5 g of ammonium acetate and 30 mL acetic acid were loaded into a three-neck refluxed overnight. The crude precipitate was washed by repeated precipitation with CH_3OH and finally recrystallized from CH_2Cl_2 and ethanol to give TPFE-Rho as a brick-red solid. The chemical structure of TPFE-Rho is showed in Fig. S1.

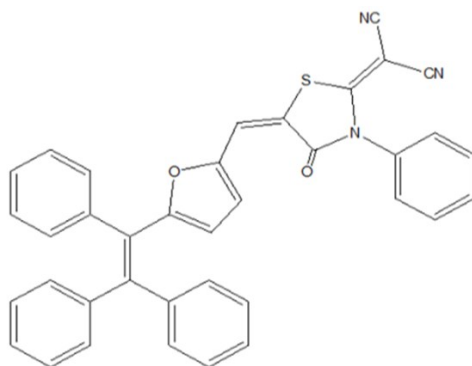


Fig. S1. Chemical structure of TPFE-Rho.

1.3 Device fabrication

1.3.1 Preparation of N-CDs/PVP thin film on a glass substrate

The glass substrates were cut into dimensions 2.0 cm \times 2.5 cm, 2.0 cm \times 4.5 cm, 2.0 cm \times 6.5 cm and polished around the edges, and then cleaned with acetone, ethanol, and deionized water by ultrasonic cleaning for 30 minutes, respectively. The N-CDs dispersed in ethanol were mixed with PVP polymer with a final concentration of PVP of 400 mg/mL. The concentration of C-dots is around 10 mg/mL. These N-CDs/polymer

were then spin-coated on glass substrates with a speed of 1000 rpm and an acceleration rate of 1500 rpm, respectively. To gain different thin film thicknesses, spin-coating with various times was employed. Finally, the thin film was dried in heating plate at 60 °C.

1.3.2 Preparation of TPFE-Rho/PMMA nanocomposite slabs

PMMA nanocomposite slabs were prepared by bulk polymerization of MMA in the presence of thermal radical initiator (AIBN). First, a blend of 20 mL of MMA monomer with 2.62 g of PMMA polymer was heated to 60 °C on a magnetic stirrer until the PMMA granules were dissolved completely. The blend was stirred vigorously for 1h at 60 °C. 0.05 g of AIBN dissolved in 5 mL MMA monomer was added to the blend of MMA and PMMA. This blend solution was stirred and heated at 60 °C for 4 h. TPFE-Rho (0.08 wt%) dispersed in 3 mL THF were added drop by drop to the blend of MMA and PMMA and the mixture was stirred for 10min (60 °C) to form a homogeneous TPFE-Rho/polymer slurry of appropriate viscosity, respectively. Then, the mixture was transferred into a casting mold. The casting mold was consisted of two glass plates with a silicone rubber between them, which were held firmly by steel clamps. The casting molds filled with N-CDs/polymer slurry were placed in the oven at 50 °C for 16 h to complete curing. Finally, the TPFE-Rho/PMMA nanocomposite slabs were cut into dimensions 2.0 cm × 2.5 cm × 0.2 cm, 2.0 cm × 4.5 cm × 0.2 cm, 2.0 cm × 6.5 cm × 0.2 cm and polished around the edges.

2. CHARACTERIZATION

UV-visible absorption spectra were recorded with a UV-Vis spectrophotometer (UV2600, Shimadzu Corp., Tokyo, Japan). Photoluminescence (PL) spectra were

measured at room temperature with a time-resolved fluorescence spectrometer (FL3-22; Jobin-Yvon, USA). Morphology of the N-CDs/PVP thin film was observed with a field-emission scanning electron microscope (FESEM, S-4800, HITACHI, Japan). PL lifetime of the TPFE-Rho in solution and in the PMMA polymer matrix was measured using a pulsed laser diode of 470 nm and Time-Correlated Single Photon Counting (TCSPC) mode in the Fluorolog-3 system. Fluorescence quantum yield (QY) was measured with UV-NIR absolute PL quantum yield spectrometer (HAMAMATSU, C13534). Photovoltaic properties of the TPFE-Rho/PMMA nanocomposite slabs and N-CDs/PVP thin film LSCs were obtained with a solar simulator (IV4112, Newport Corp., Irvine, CA, USA) at an intensity of $100 \text{ mW}\cdot\text{cm}^{-2}$ (1 sun).

3. RESULTS

Table S1 Photovoltaic parameters of the tandem LSCs.

Luminophore	LSC size (cm^3)	J_{SC} (mA/cm^2)	V_{OC} (V)	Fill Factor (%)	G	η_{opt} (%)	η (%)	Tandem LSC η (%)
N-CDs	$2.5 \times 2.0 \times 0.1$	5.78	0.49	62.07	5.5	2.28	1.76	4.06
TPFE-Rho	$2.5 \times 2.0 \times 0.2$	6.88	0.53	63.18	2.8	2.92	2.30	
N-CDs	$4.5 \times 2.0 \times 0.1$	5.70	0.48	61.89	6.9	2.05	1.69	3.89
TPFE-Rho	$4.5 \times 2.0 \times 0.2$	6.87	0.51	62.89	3.5	2.75	2.20	
N-CDs	$6.5 \times 2.0 \times 0.1$	5.18	0.47	61.88	7.6	1.73	1.51	3.55
TPFE-Rho	$6.5 \times 2.0 \times 0.2$	6.85	0.48	62.20	3.8	2.31	2.04	

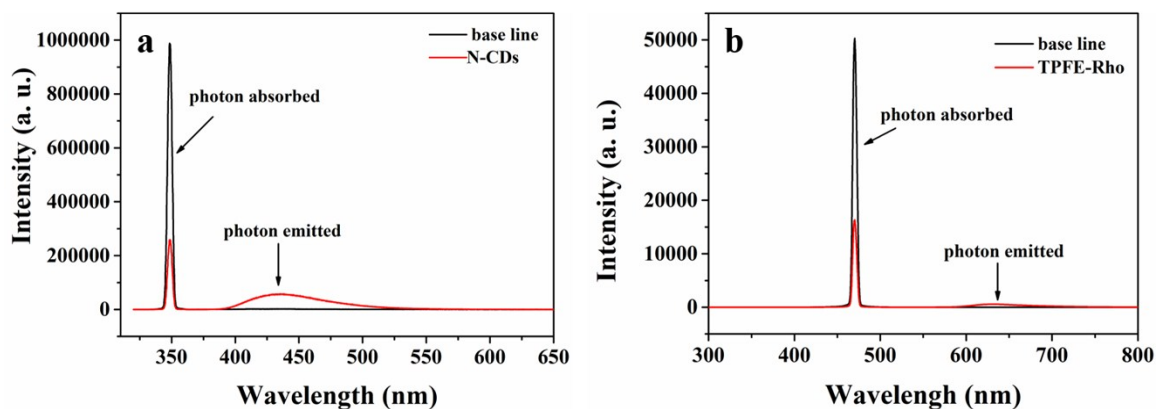


Fig. S2. Experimental data of fluorescence quantum yield measurement for N-CDs (a) and TPFE-Rho (b).

The fluorescence quantum yield (QY) can be determined by the ratio of the integrated areas between the emission and excitation ranges. The absolute QYs of N-CDs and organic nanoparticle TPFE-Rho are $\Phi_{\text{N-CDs}} = 79.65\%$ and $\Phi_{\text{TPFE-Rho}} = 34.88\%$, respectively.

Reference

S1. Li, Y. X.; Miao, P.; Zhou, W.; Gong, X.; Zhao, X. J., N-doped carbon-dots for luminescent solar concentrators. *J. Mater. Chem. A* **2017**, *5* (40), 21452-21459.

S2. Xia, Q.; Chen, Z. K.; Yu, Z. Q.; Wang, L.; Qu, J. Q.; Liu, R. Y., Aggregation-Induced Emission-Active Near-Infrared Fluorescent Organic Nanoparticles for Noninvasive Long-Term Monitoring of Tumor Growth. *ACS Appl. Mater. Inter.* **2018**, *10* (20), 17081-17088.