Supporting Information

Nitrogen-Doped Graphene Quantum Dots for 80% Photoluminescence Quantum Yield for Inorganic γ-$CsPbI_3$ Perovskite Solar Cells with Efficiency beyond 16%

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Experimental

Materials preparation: All materials used here were purchased from Sigma-Aldrich and were used as received without further purification. 2.2 mm FTO-coated glass and PET were used as the substrates.

N-GQDs synthesis: 0.2 g o-phenylenediamine (98%) was dissolved in 20 mL alcohol (100 mg/mL, 96%). The solution was transferred into a 25 mL Teflon-lined autoclave and heated at 180 ℃ for 24 h. The obtained light yellow solution was added to 50 mL water for dialysis (100 Da) for 3 days to produce 5 wt% N-GQDs.

Precursor solution preparation: The γ-$CsPbI_3$ precursor solutions were prepared used $HPbI_3$ (0.88 g, 99.99%), CsI (0.36 g, 99.999%) and PEAI (10 mg) dissolved in 2 ml DMF/DMSO (v/v 9:1, 99.8%) in N_2 atmosphere in a glovebox with active stirring for 24 h.

HTL solution preparation: A solution was prepared by dissolving PTAA (36 mg, M_n=26,000, M_w=63,000), a sulfonyl imide (Li-TFSI, 99%, 22 μL, 520 mg Li-TFSI in 1 mL acetonitrile, 99.9%), and tert-butylpyridine (TBP, 36 μL, 98%) in 1 mL of CB solution (99.8%).

Preparation of TiO_2-blocking layer: A 25 × 25 mm^2 piece of FTO-coated glass was washed, dried and treated with O_2 plasma. The clean substrate was immersed in a 40 mM $TiCl_4$ (99.6%) aqueous solution for 30 min at 70 ℃ and washed with distilled water and ethanol, followed by annealing at 200 ℃ for 30 min in air to form a compact n-type blocking layer of TiO_2. Afterward, the N-GQDs EDS layer and polymethyl methacrylate (PMMA layer) was fabricated in turns on the glass side by spin-coating the N-GQDs solution at 4500 rpm for different times.

Growth of the γ-$CsPbI_3$ film: The γ-$CsPbI_3$ layer was fabricated via one-step spin-coating of the solution onto O_2-plasma-treated substrate at 1000 rpm for 10 s. The speed was then increased to 3500 rpm and maintained for 40 s. Finally, the films were annealed at 150 ℃ for 2 h.

Assembly of the solar cells: An HTL film was prepared by spin-coating the HTL solution onto the γ-$CsPbI_3$ film at 5000 rpm for 30 s. Immediately after, a 70-nm-thick gold electrode was thermally evaporated onto the HTL-coated film.

TEM, AFM and SEM: The TEM image was obtained with a FEI Tecnai T20. AFM (Dimension ICON) measurements used the tapping mode. The film surface morphology was characterized by FESEM (SEM, Jeol SU-8020).

Absorbance and PL: Absorbance spectra were collected using a Shimadzu UV-3600. The PL spectra were measured using a PicoQuant FluoTime 300. The source light was a xenon short arc lamp.

XRD and XPS: XRD patterns of the samples were obtained using a Bruker D8 GADDS Diffractometer with the Cu Kα line. The XPS measurements were performed in a VG ESCALAB MK2 system.

J-V and EQE: The J-V measurement was performed using a solar simulator

(SS-F5-3A, Enlitech) with the AM 1.5G spectra, whose intensity was calibrated at 100 mW/cm^2 using a certified standard silicon solar cell (SRC-2020, Enlitech). The measurements were made in the reverse scan mode (from V_{OC} to I_{SC}) and forward scan mode (from I_{SC} to V_{OC}) with a scan rate of 30 mV/s. The external quantum efficiency (EQE) data were obtained by using the solar-cell spectral-response measurement system (QE-R301，Enlitech).

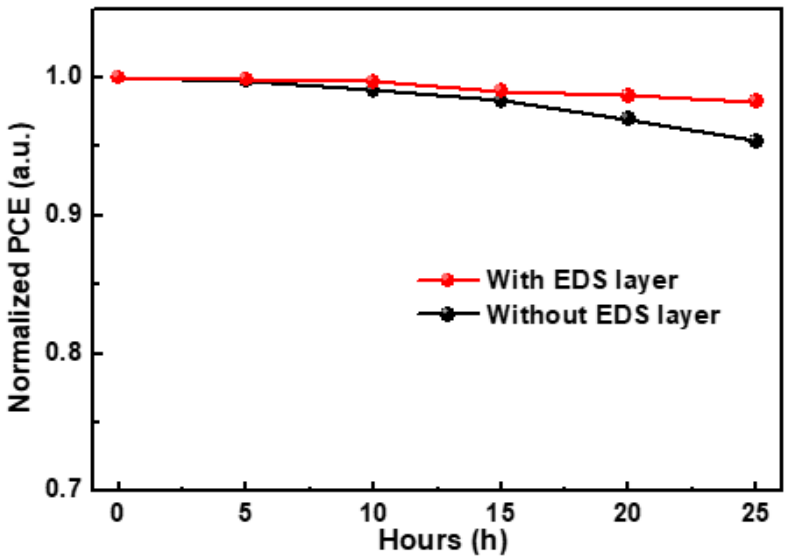

Figure S1. Results of the aging test on the PSCs with and without EDS layer under continuous UV irradiation (5 mW/cm^2) at N_2 atmosphere for 24 h.