**Supplementary material**

**Defect passivation and electrical conductivity enhancement in perovskite solar cells using functionalized graphene quantum dots**

Yichuan Rui1,\*, Zuoming Jin1, Xinyi Fan1, Weitao Li2, Bin Li1, Tianpeng Li1,3, Yuanqiang Wang1, Liang Wang2,\* Jia Liang3,\*

1 College of Chemistry and Chemical Engineering, Shanghai University of engineering Science, Shanghai 201620, China.

2 Institute of Nanochemistry and Nanobiology, School of Environmental and Chemical Engineering, Shanghai University, Shanghai 200444, China.

3 Department of Materials Science, Fudan University, 220 Handan Road, Shanghai 200433, China

**Characterization.** Field emission scanning electron microscope (FESEM, Model SU8010, Hitachi) was used to perform the perovskite films and devices morphological properties. PL spectra of GQDs in different excitation wavelengths was measured by FS-5 spectrophotometer (URT210-C, Edinburgh). X-ray photoelectron spectroscopy (XPS) was applied by Escalab 250 Xi. The Confocal laser scanning microscope (CLSM) was carried out with Leica TCS SP5 with the excitation wavelength of 458 nm. Steady state photoluminescence (PL) spectra and time-resolved photoluminescence (TRPL) spectra were measured at 480 nm excitation wavelength with FLS920 transient optical spectrometer (Edinburgh Instruments, UK). Thermalgravimetric analysis (TGA) of the perovskite films were carried out on a NETZSCH STA449C thermal analyzer. Simulated sunlight was provided by Model 96160 Newport company and the light intensity was adjusted to 100 mW cm-2 by a NREL-calibrated silicon solar cell. Photocurrent density-voltage (*J-V*) curves of the perovskite solar cells were performed by using Keithley 2400 Source Meter. The incident-photon-to-current conversion efficiency (IPCE) spectra were obtained and the wavelength was set from 300 to 900 nm (Newport). Electrochemical impedance spectroscopy (EIS) was performed using Zahner-elecktrik analyzer (Germany) at bias of 0.9 V and frequency range of 100 mHz to 100 kHz under dark condition.



**Figure S1** PL spectra of GQDs under different excitation wavelength.



**Figure S2** Fourier transform infrared (FT-IR) spectrum of the dried GQDs powder.



**Figure S3** Top-view SEM image of the MAPbI3/0.1GQDs perovskite film.



**Figure S4** *J–V* curve of the PSCs based on the MAPbI3/0.0005 GQDs.

**Table S1** The best photovoltaic parameters of the PSCs based on the MAPbI3 and MAPbI3/xGQDs (x = 0.0005, 0.001, 0.005, and 0.01) perovskite films.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Sample | *V*oc (V) | *J*sc (mA cm-2) | FF (%) | PCE (%) |
| MAPbI3 | 1.072 | 22.25 | 67.3 | 16.05 |
| MAPbI3/0.0005GQDs | 1.134 | 22.70 | 71.0 | 18.28 |
| MAPbI3/0.001GQDs | 1.158 | 23.36 | 72.4 | 19.59 |
| MAPbI3/0.005GQDs | 1.150 | 22.90 | 70.3 | 18.51 |
| MAPbI3/0.01GQDs | 1.122 | 22.49 | 70.4 | 17.78 |



**Figure S5** Schematic diagram of the energy level optimization by GQDs.

**Table S2** The trap-filled limit voltage, trap-state density and hole mobility determined from SCLC curves.

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Samples | MAPbI3 | MAPbI3/0.001GQDs | MAPbI3/0.005GQDs | MAPbI3/0.01GQDs |
| *V*TFL [V] | 1.11 | 0.50 | 0.36 | 0.84 |
| *n*t [cm-3] | 1.57×1016 | 7.08×1015 | 5.10×1015 | 1.19×1016 |
| *μ* [cm2 V-1 s-1] | 3.84×10-3 | 8.56×10-3 | 4.40×10-3 | 3.50×10-3 |



**Figure S6** The evolution of contact angles on various perovskite films: (a) MAPbI3, (b) MAPbI3/0.001 GQDs, (c) MAPbI3/0.005 GQDs, and (d) MAPbI3/0.01 GQDs samples, respectively.