

## Supporting Information

### **Synergetic Effects of Solution-Processible Fluorinated Graphene and PEDOT as a Hole-Transporting Layer for Highly Efficient and Stable Normal-Structure Perovskite Solar Cells**

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## Experimental section

### Preparation of FrGO

Graphene oxides (GOs) were prepared via the previously reported method.<sup>23</sup> For reduction process, the resultant GOs were dispersed in 100 ml of deionized (DI) water by using an ultrasonic bath with a concentration of 4 mg ml<sup>-1</sup>. The 2 ml of reductant (4-(trifluoromethyl)phenyl hydrazine, Sigma-Aldrich) was added into the GO colloidal solution, and stirred for 6 h. Subsequently, the colloidal solution was filtered by using a vacuum filter, and then, washed by alcohol. The filtered materials were dried in an oven at 60 °C. Finally, the FrGO was dispersed in 2-propanol at a concentration of 1 mg ml<sup>-1</sup> for device-fabrication.

### PeSC fabrication and characterization

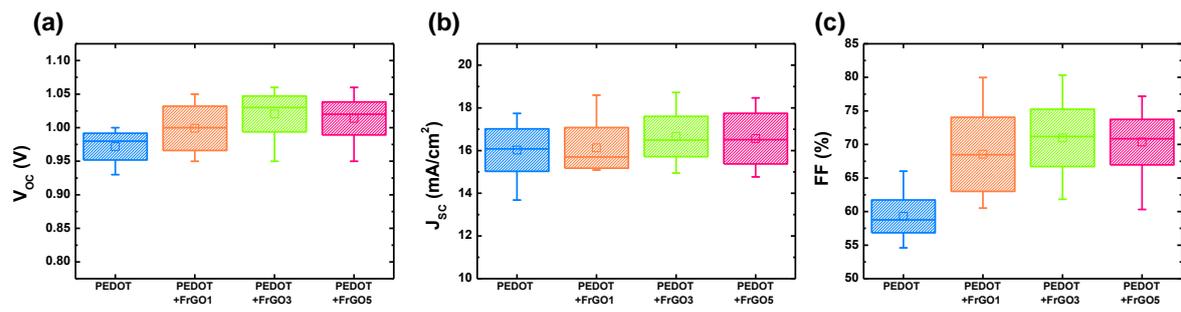
The configuration of the PeSCs studied in this report was ITO/ZnO/C60/MAPbI<sub>3</sub>/PEDOT/FrGO/MoO<sub>3</sub>/Ag. The pre-patterned ITOs (10 Ohm sq<sup>-1</sup>, Samsung Corning) on glass-substrates were sequentially cleaned with acetone, deionized water, and 2-propanol under an ultrasonic bath for 20 min, respectively, and dried in an oven at 80 °C for 10 min. Then, the substrates were exposed in UV-ozone for 30 min. For ZnO layer, zinc oxide nanopowder (ZnO nanopowder, < 100 nm particle size, Sigma-Aldrich) dissolved in ammonium hydroxide solution (NH<sub>4</sub>OH, 50% v/v aqn. soln., Alfa Aesar) at a concentration of 8 mg ml<sup>-1</sup> was spin-coated on the ITO-coated glass at 5000 rpm for 40 s, and subsequently dried at 200 °C for 20 min in air. Then, C60 (Fullerene, 99.5%, Nano-C) dissolved in 1,2-dichlorobenzene (15 mg ml<sup>-1</sup>) was spin-coated on the ITO/ZnO substrates at 2000 rpm for 40 s, and annealed at 100 °C for 10 min in an N<sub>2</sub> glove box. Next, for the perovskite layer, PbI<sub>2</sub> (Lead(II) iodide, 99.9985%, Alfa Aesar) of 0.759 g and MAI (Dyesol) of 0.262 g were dissolved in DMF (anhydrous 99.8%, Sigma-Aldrich) solvent of 1.612 ml, and

then, 120 ml of N-cyclohexyl-2-pyrrolidone (CHP, Sigma-Aldrich) was added into the solution. The prepared perovskite precursor solution was spin-coated on the ZnO/C60 at 4000 rpm for 40 s, followed by annealing at 100 °C for 4 min in an N<sub>2</sub> glove box. For the HTLs, PEDOT (Clevios HTL Solar 3) and FrGO solutions were spin-coated on the active-layer at 5000 rpm for 40 s, and then dried at 100 °C for 5 min in an N<sub>2</sub> glove box, respectively. Finally, the MoO<sub>3</sub> (2.5 nm)/Ag (80 nm) electrodes were deposited via thermal evaporator with a shadow mask to define an active-area of 4.64 mm<sup>2</sup>.

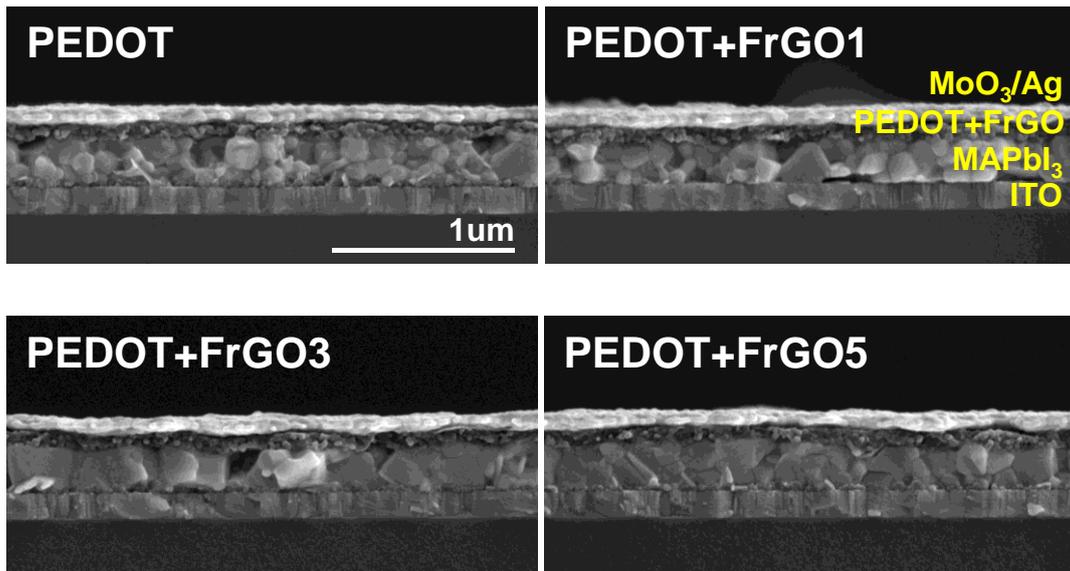
### **Device measurement and characterization**

The photocurrent density-voltage (J-V) characteristics were measured by Keithley 2400 source measurement under AM 1.5 G illumination (100 mW cm<sup>-2</sup>) using a solar simulator. Both external and internal quantum efficiency (EQE and IQE) of PeSCs were analyzed using a certified IPCE measurement system (IQE-200, Oriel Instruments). The integrated values of the J<sub>SC</sub> obtained from the IPCE data are in close agreement (within nearly 9%) with the values obtained from the J-V curves. To analyze interface characteristics, the impedance measurement of PeSCs were performed in white light condition by using Solartron 1260 Impedance/gain-phase analyzer, with frequency range of 100,000 Hz to 0.1 Hz and AC oscillating voltage of 0.90 V. A Scribner ZView 2 software was used as the fitting program of the spectra. X-ray photoelectron spectroscopy (XPS) and ultraviolet photoelectron spectroscopy (UPS) (AXIS-NOVA, Kratos Inc.) were carried out with a monochromatic Al-K $\alpha$  (1486.6 eV) for XPS and a He 1 (h $\nu$  = 21.2 eV) for UPS, respectively. The surface morphology and surface potential of the HTLs were detected by atomic force microscopy (AFM) and scanning Kelvin probe microscopy (SKPM) (Dimension 3100, Veeco), respectively. In order to compare optical transmittance according to spin-coating cycles of FrGO solution, UV-vis

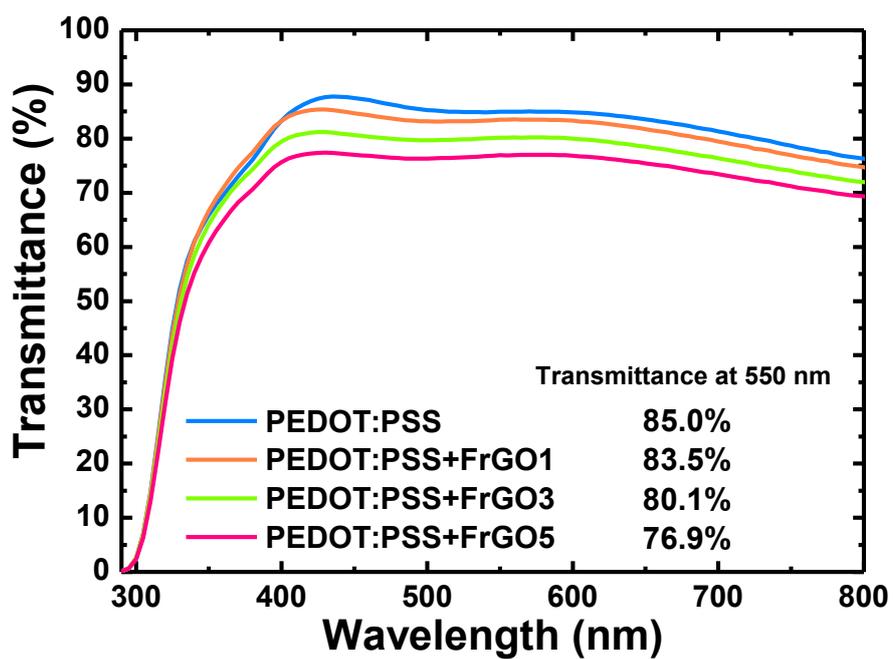
(LAMBDA 750 UV/Vis/NIR Spectrophotometer, PerkinElmer Inc.) was used. The contact-angles of water-droplets on the HTLs were measured by Phoenix 300 (SEO Inc.). Scanning electron microscopy (SEM, Quanta 3D-FEG/FEI) was used to obtain cross-sectional image of the PeSCs.



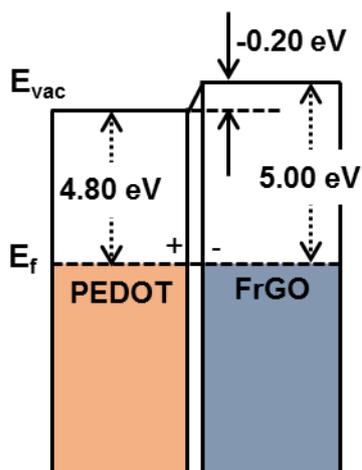
**Fig. S1.** Statistical data of (a)  $V_{oc}$ , (b)  $J_{sc}$ , and (c) FF for the PeSCs with different HTLs.



**Fig. S2.** (a) Cross-sectional SEM images of the different PeSCs.



**Fig. S3.** (a) Optical transmittance of different films on glass substrates.



**Fig. S4.** (a) Schematic energy-level diagram between PEDOT and FrGO.