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Supplementary Information

Extending the environmental lifetime of unpackaged perovskite solar cells through interfacial design

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Experimental Section

Materials: PEDOT:PSS and $PC_{61}BM$ (99.5%) were purchased from Clevious and Solenne BV, respectively. PbI_2 and Methylammonium iodide (CH_3NH_3I) were purchased from Sigma-Aldrich and Dyenamo, respectively. Oxo-functionalized graphene (oxo-G_1) was synthesized using graphite crystals (grade *3061*) from Asbury Carbons Inc. Potassium permanganate, sulfuric acid, and hydrogen peroxide were purchased from Sigma-Aldrich®, double distilled water from Carl Roth®. For centrifugation, we used a Sigma *4K15* centrifuge with 200 ml plastic beakers. For ultrasonication, we used a *UW3200* sonotrode (Bandelin) equipped with a flat titan tip (working frequency: 20 kHz) and a maximum output power of 200 W. Oxo-G_1 was synthesized as described in literature before, but with minor changes[31, 40]. Graphite crystals (1 g, 83.33 mmol) were stirred in cold conc. H_2SO_4 (96%, 24 mL, < 10 °C). $KMnO_4$ (2 g, 12.66 mmol) was added over 4 h and stirred for further 16 h. Then, diluted H_2SO_4 (20%, 20 mL) was added continuously within 4 h followed by continuous addition of water (100 mL). Subsequently, aqueous hydrogen peroxide (5%, 50 mL) was added to reduce insoluble manganese species. At all time, the process was performed at temperatures <10 °C by the use of a refrigerating coil. Obtained oxo-G_1 was purified through repetitive centrifugation (6 times, 1500 RCF), followed by delamination through pulsed ultrasonication for 4 minutes (20 W; 2 s on/2 s off). The dispersion was centrifuged at 1500 RCF (3 times, 30 min) to remove non-monolayer particles. The supernatant was again centrifuged at 13000 RCF (one time, 45 min) to remove minor amounts of smallest particles. Finally, oxo-G_1 suspension was diluted 1:1 (vol:vol) with isopropanol and a stable yellowish oxo-G_1 suspension was obtained (0.25 mg/ml in water/isopropanol). Analytical data can be found in the literature [31, 40].

Device Fabrication and Characterization: Patterned FTO substrates were successively ultrasonic cleaned with toluene, acetone and isopropanol. Then, the cleaned FTO substrates were spin-coated with PEDOT:PSS and followed by annealing at 140 °C for 10 min in ambient air. Oxo-G_1 suspension was spin-coated on FTO substrates and annealed at 120 °C for 10 min in ambient atmosphere. PbI_2 and CH_3NH_3I were mixed in a mixture of DMF and DMSO (2:1 v/v) with molar ratio of 1:1 at a concentration of 40 wt.%. The solution was spin-coated onto FTO/PEDOT substrates at 4000 rpm for 35 sec. Then, the substrates were annealed at 100ºC for 10 minutes. After that, 20 mg/ml PCBM in chlorobenzene solution was coated onto the perovskite layer at 2000 rpm for 30 sec. Subsequently, ZnO solution (nanoparticle suspensions, provided by NanoGrade) was deposited by spin-coating at 3000 rpm. Finally, 150 nm Al were thermally evaporated on ZnO through a shadow mask under 10^{-6} mbar to form an active area of 10.4 mm^2.

J-V curves were measured using a source measurement unit from BoTest with a Newport Sol1A solar simulator at a scan rate of 100 mV/s. The EQE spectra were measured using an Enli Technology (Taiwan) EQE measurement system (QE-R). The morphology data was obtained by AFM (Veeco Model D3100, tapping mode). White light LEDs (BRIDGELUX-BXRA-30E0800-B-00) are used for photodegradation of the perovskite solar cells. In order to reduce the effect of heat on the degradation of perovskite solar cell, continuous illumination was provided by a white light LED with a light intensity of ~0.5 suns. ATR-FTIR spectra were carried out by using a Vertex 70 from Bruker optics with a DTGS detector.

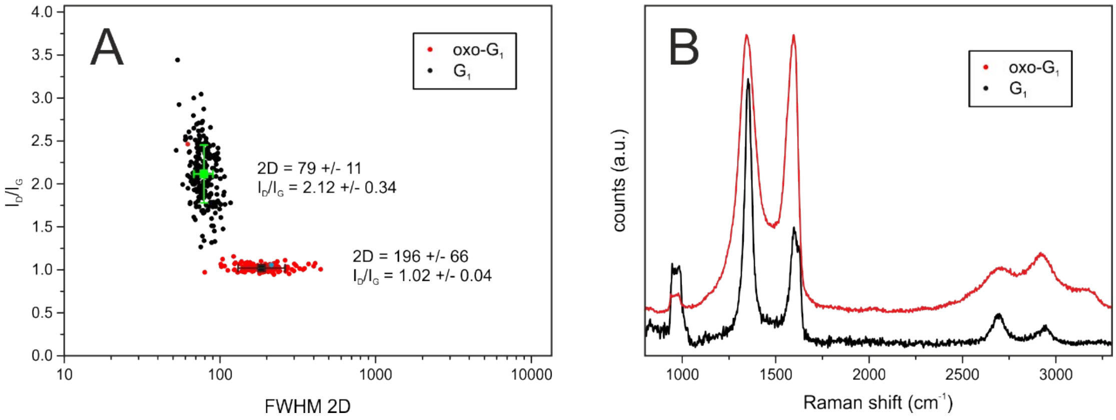

Figure S1. (A) I_D/I_G ratio of oxo-G$_1$ and G$_1$ spectra vs. FWHM of the 2D peak, (B) Raman spectrum of oxo-G$_1$ and G$_1$.

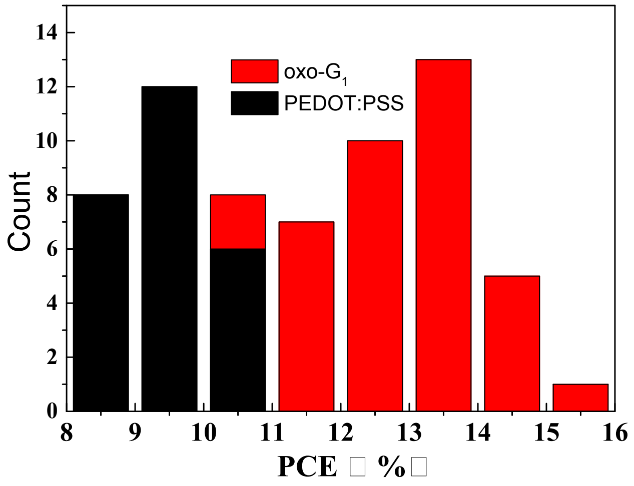

Figure S2. Performance distribution of oxo-G$_1$ devices and PEDOT:PSS devices.

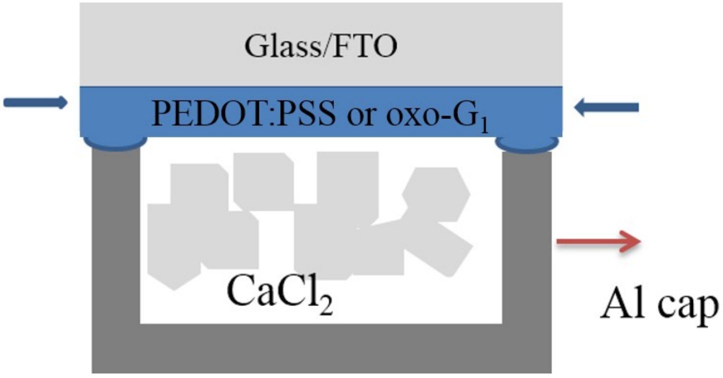

Figure S3. Water vapor transmission measurement at 85 ℃/85% RH.

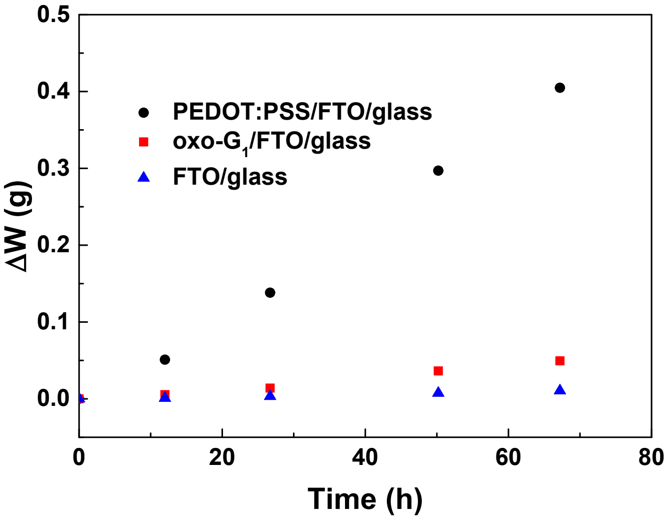

Figure S4. Increase in weight of PEDOT:PSS/FTO/glass, oxo-G_1/FTO/glass and FTO/glass over time at 85 ℃/85% RH.