Supporting Information

A Simple Perylene Derivative as a Solution−Processable Cathode Interlayer for Perovskite Solar Cells with Enhanced Efficiency and Stability

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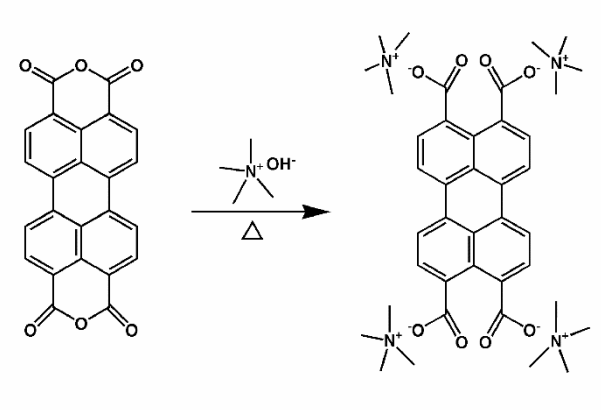

Scheme S1. Synthetic route of TMA-PTC.

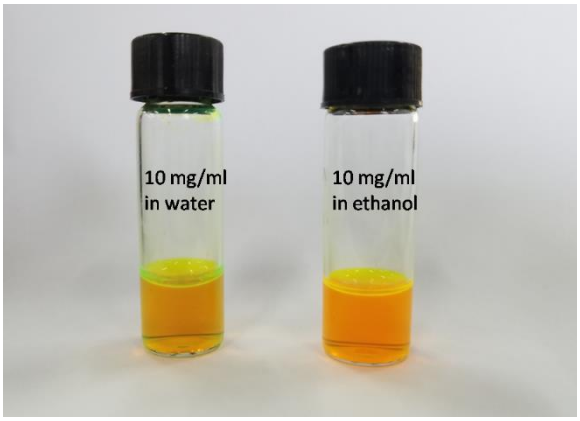

Figure S1. Photos of TMA-PTC water and ethanol solutions at the concentration of 10 mg/ml, respectively.

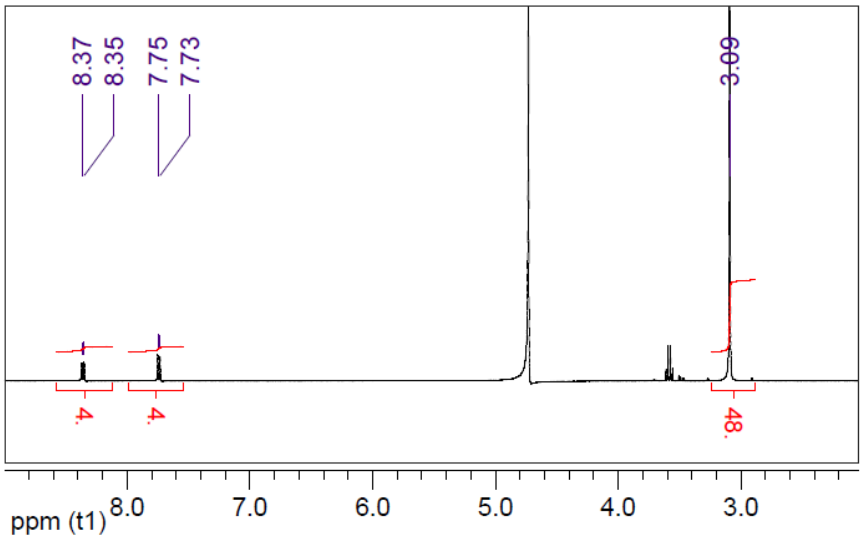

Figure S2. ^{1}H NMR spectrum of TMA-PTC.

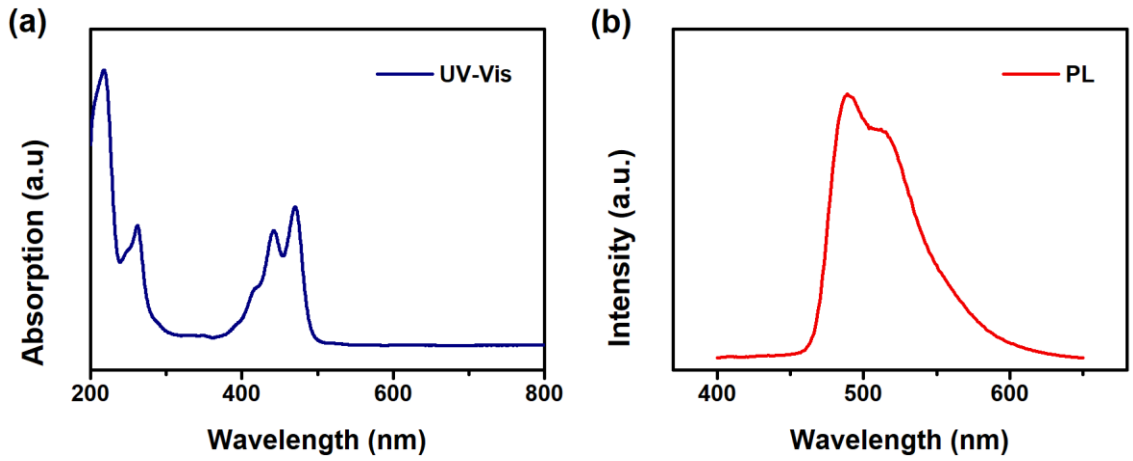

Figure S3. (a) UV-Vis absorption and (b) PL spectra of TMA-PTC in ethanol (0.5 mg/mL).

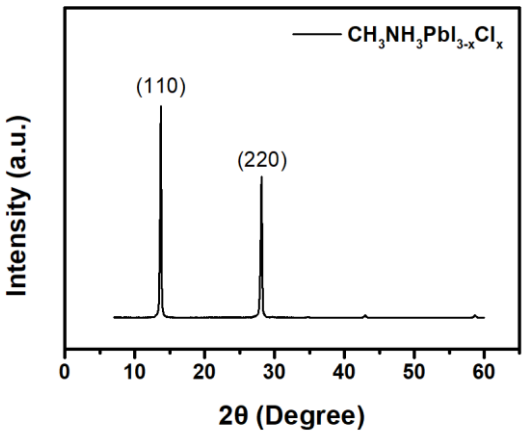

Figure S4. XRD patterns of the perovskite film prepared from the mixture of PbI_2:$PbCl_2$:CH_3NH_3I and annealed at 80 ℃ for 2 hours.

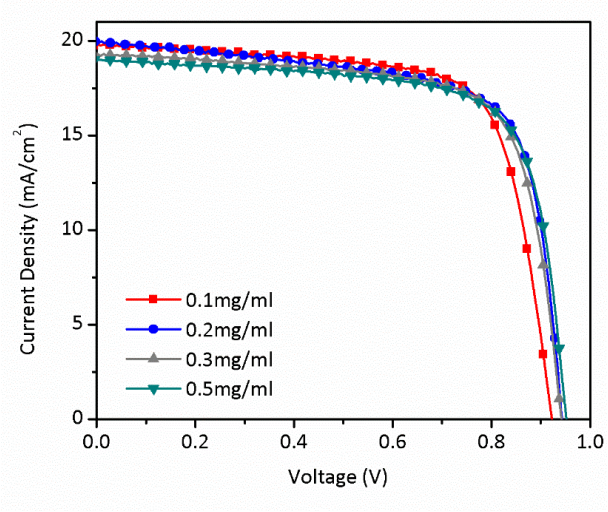

Figure S5. *J-V* curves of PeSCs with TMA-PTC CIL prepared from the solutions with varied concentrations.

Table S1. Photovoltaic parameters of PeSCs with TMA-PTC CIL with varied concentrations.

concentration	V_{oc}(V)	FF (%)	J_{SC}(mA/cm^2)	PCE (%)
0.1mg/ml	0.925	71.74	19.82	13.14
0.2mg/ml	0.940	70.94	19.99	13.33
0.3mg/ml	0.944	72.22	19.33	13.18
0.5mg/ml	0.951	72.77	19.01	13.15

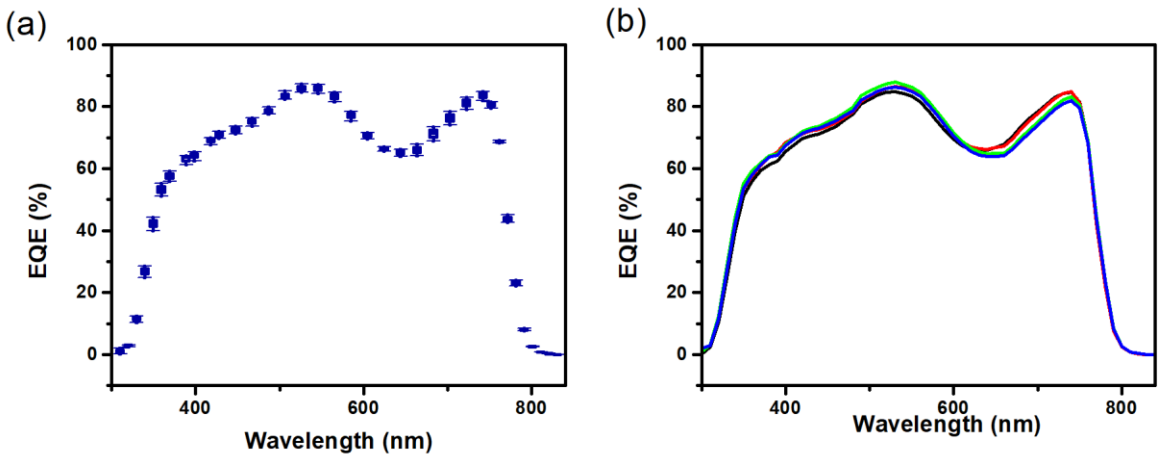

Figure S6. (a) EQE spectrum of a typical perovskite solar cells with TMA-PTC CIL, and (b) EQE spectra of four individual perovskite solar cells with TMA-PTC CIL.

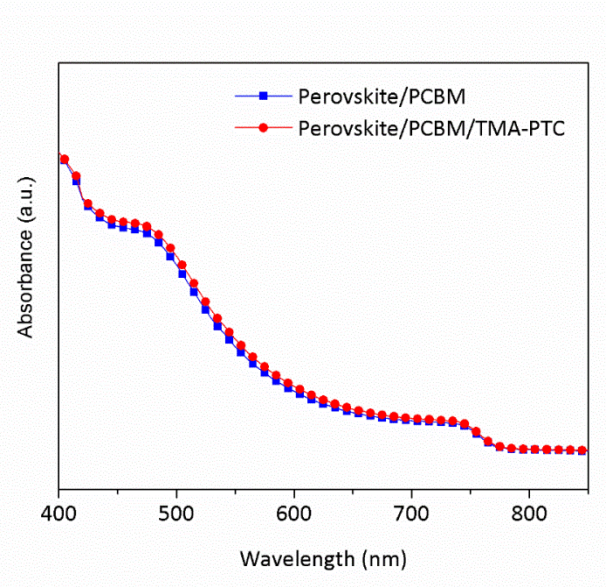

Figure S7. UV-Vis absorption spectra of perovskite films coated with $PC_{61}BM$ and $PC_{61}BM$/TMA-PTC, respectively.

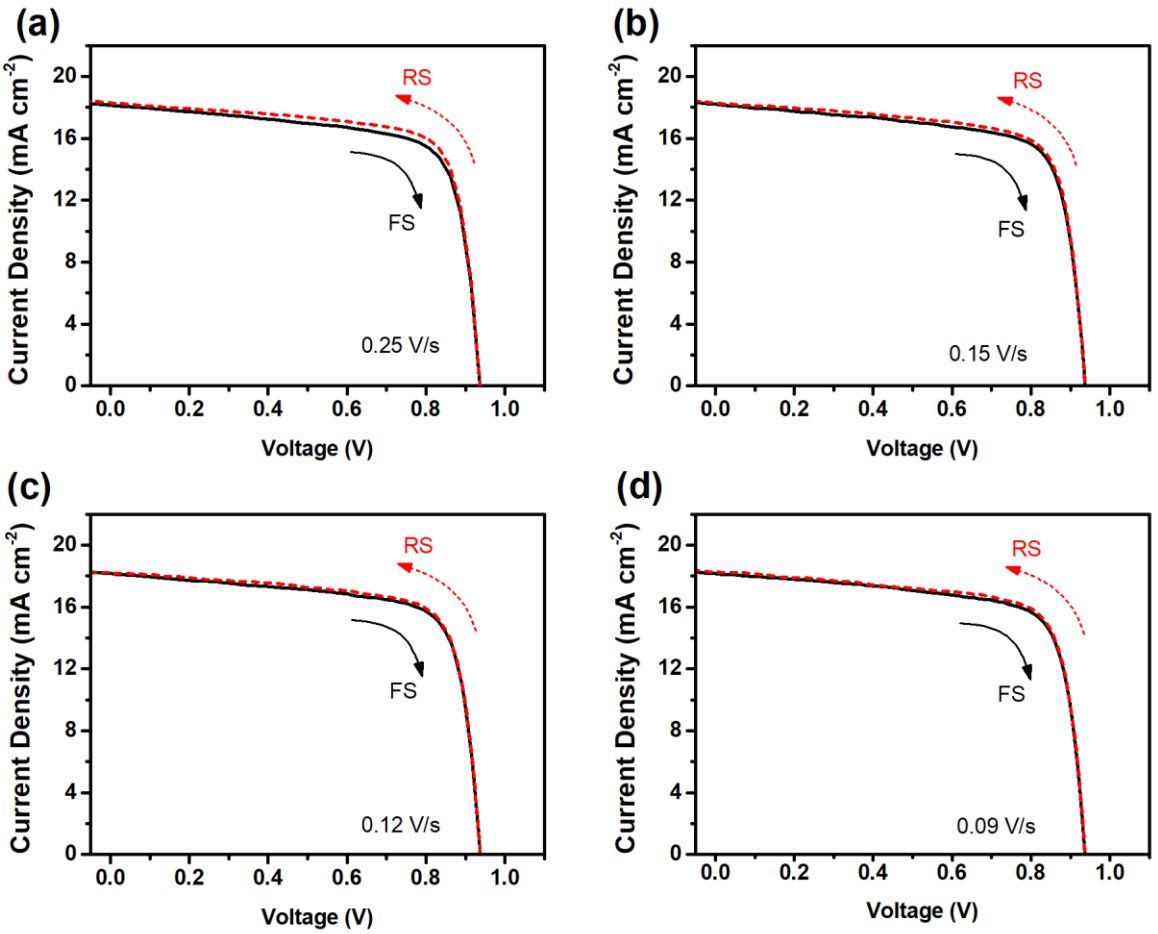

Figure S8. *J*−*V* curves of PeSCs (not the best performance device) with TMA−PTC as the CIL under both forward (FS) and reverse (RS) scans with varied scan speeds (i.e., (a) 0.25 V/s, (b) 0.15 V/s, (c) 0.12 V/s, (d) 0.09 V/s) in 1 sun illumination.

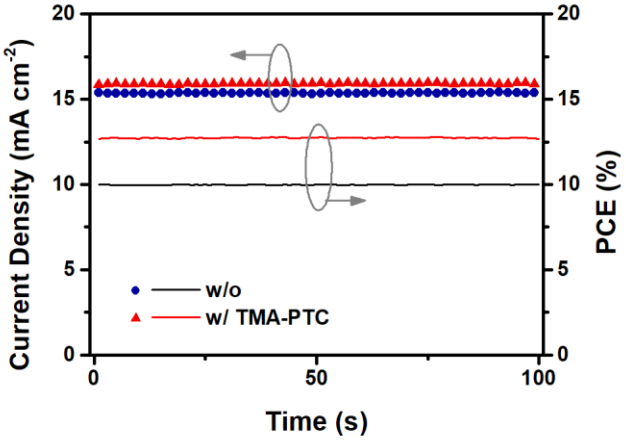

Figure S9. Steady-state measurement of J_{SC} and PCE for the PeSCs (not the best performance devices) with and withou TMA-PTC CIL at the corresponding maximum power output points (i.e., at 0.80 V and 0.65 V, respectively).

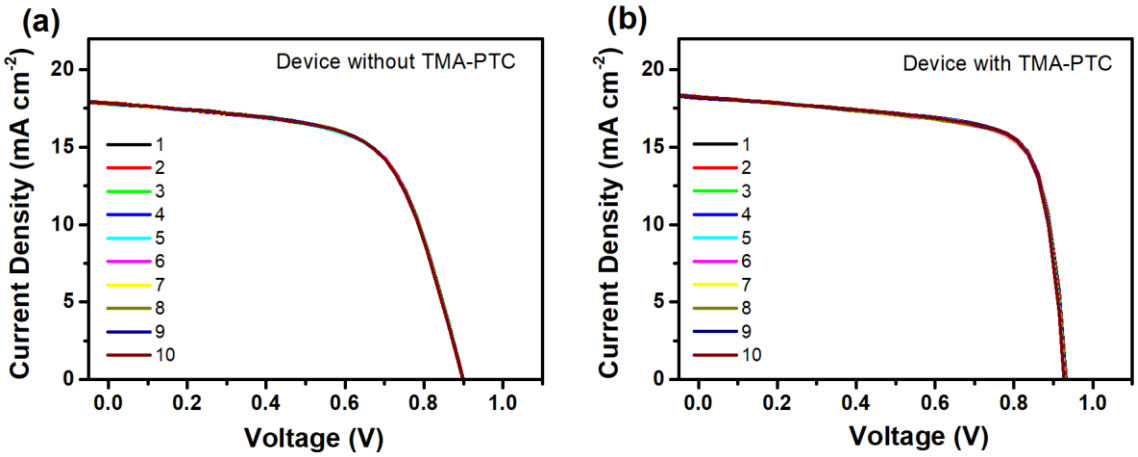

Figure S10. $J-V$ curves of PeSCs (not the best performance devices) (a) with and (b) without TMA-PTC CIL measured at repeated scans of 10 times in 1 sun illumination.

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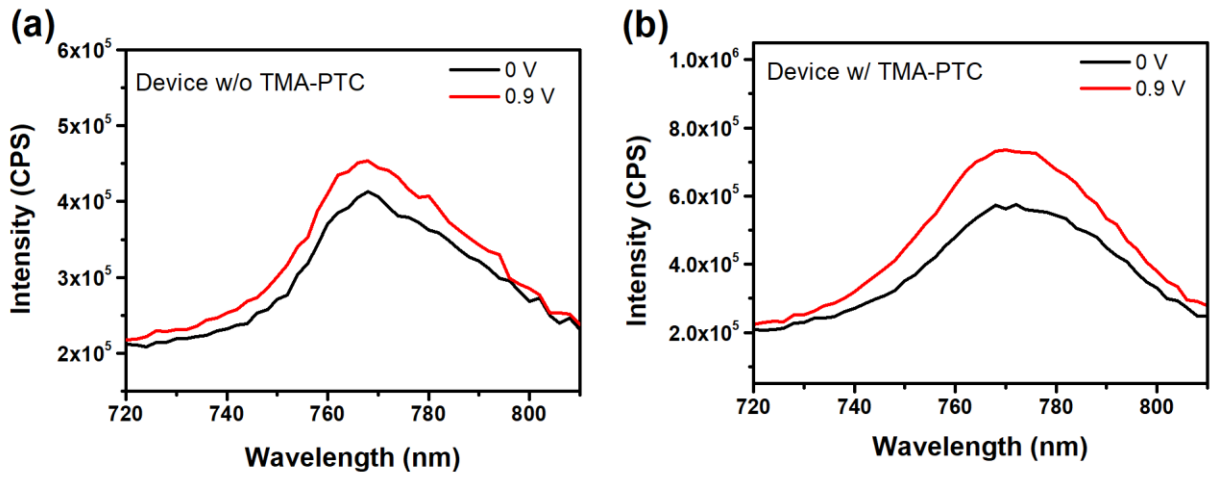

Figure S11. (a) PL of the device without TMA-PTC at 0 V and 0.9 V; (b) PL of the device with TMA-PTC at 0 V and 0.9 V.

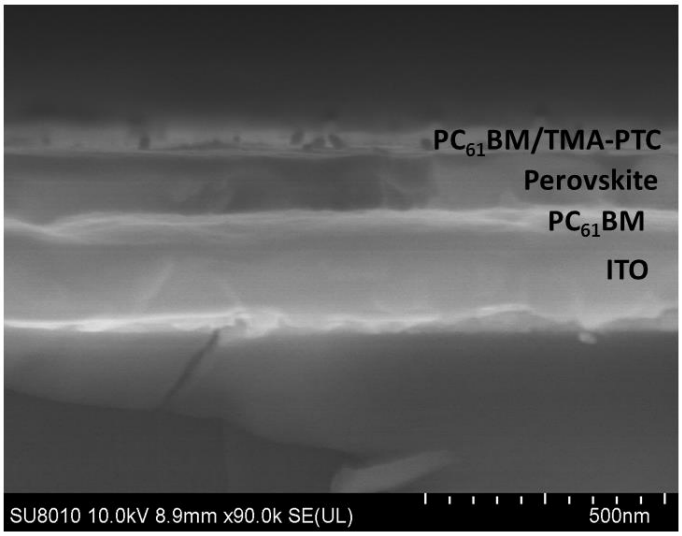

Figure S12. Cross sectional SEM image of ITO/$PC_{61}BM$/Pervoskite/$PC_{61}BM$/TMA−PTC stacked films in electron-only device.

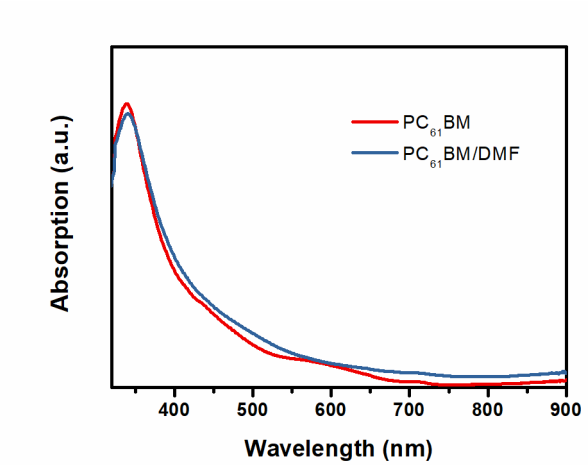

Figure S13. Absorption spectra of $PC_{61}BM$ films before and after spin-coated with DMF solvent.

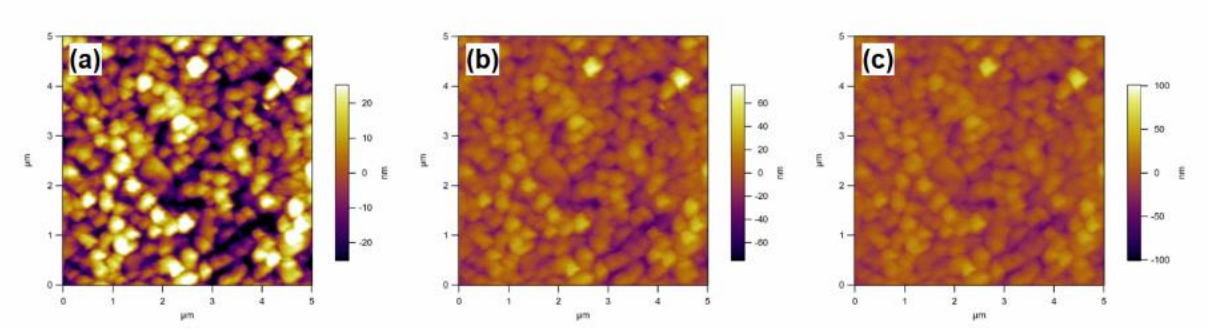

Figure S14. AFM images of pristine perovskite films with different color contrast: (a) 50 nm; (b) 100 nm; (c) 200 nm.

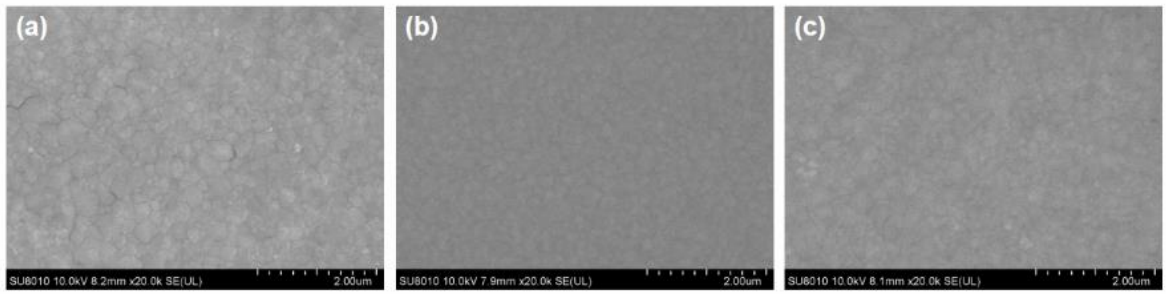

Figure S15. Top-view SEM images of (a) pristine perovskite, (b) perovskite/$PC_{61}BM$ and (c) perovskite/$PC_{61}BM$/TMA-PTC layers.

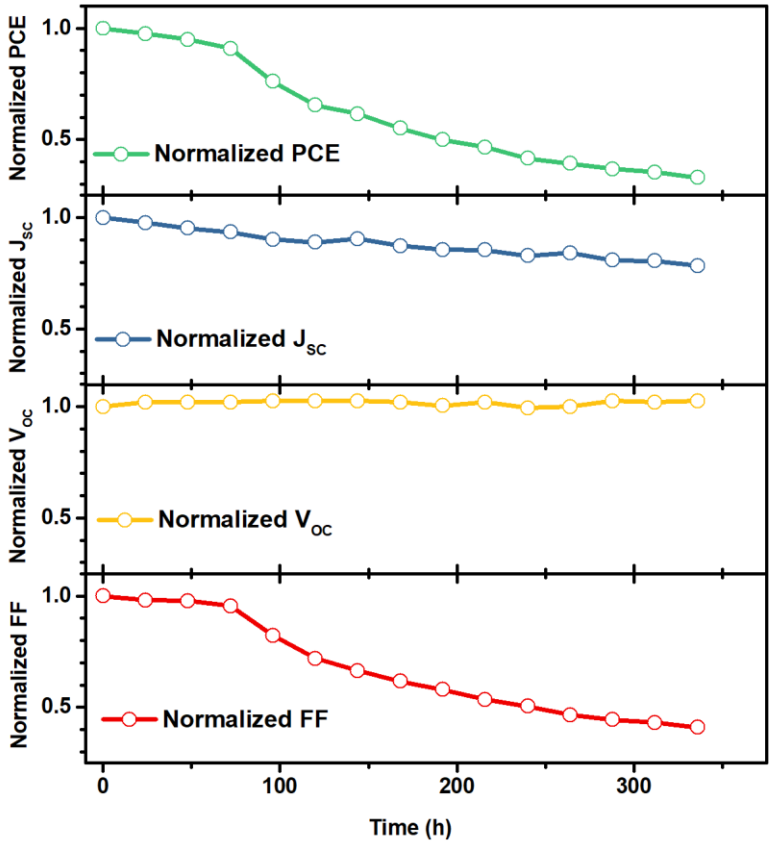

Figure S16. Photovoltaic parameters of perovskite solar cell without TMA-PTC decayed as a function of exposion time in the air.

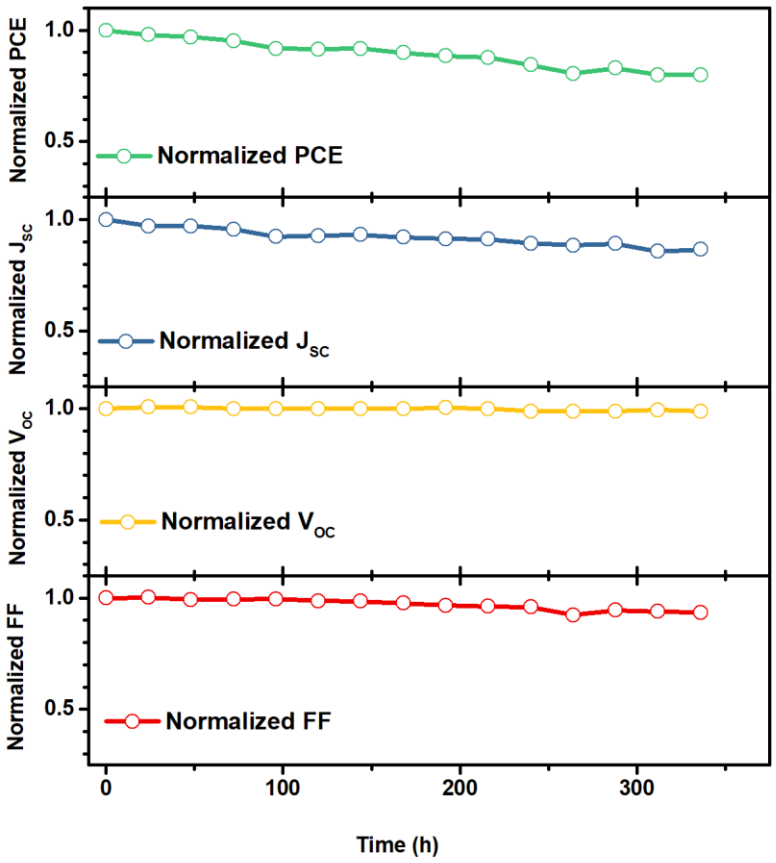

Figure S17. Photovoltaic parameters of perovskite solar cell with TMA-PTC decayed as a function of exposion time in the air.

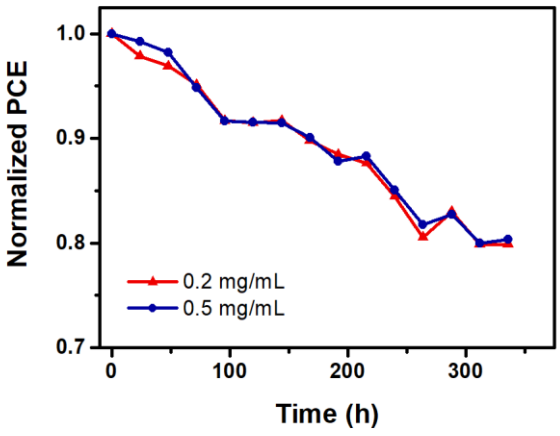

Figure S18. PCE decay of PeSCs treated with 0.2 and 0.5 mg/mL TMA-PTC decayed as a function of exposion time in the air.

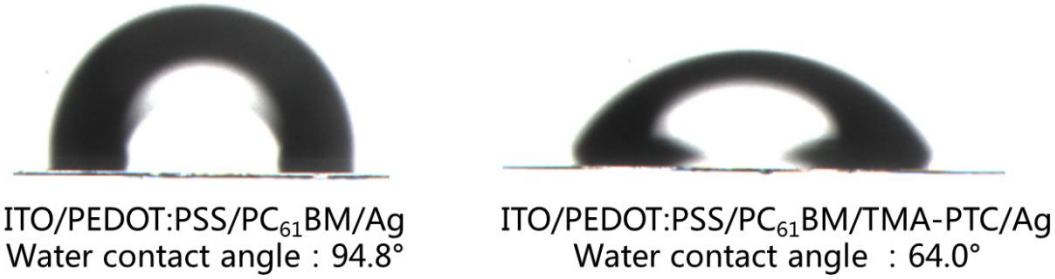

Figure S19. Measured water contact angle for the $PC_{61}BM$ films with and without TMA-PTC CIL.

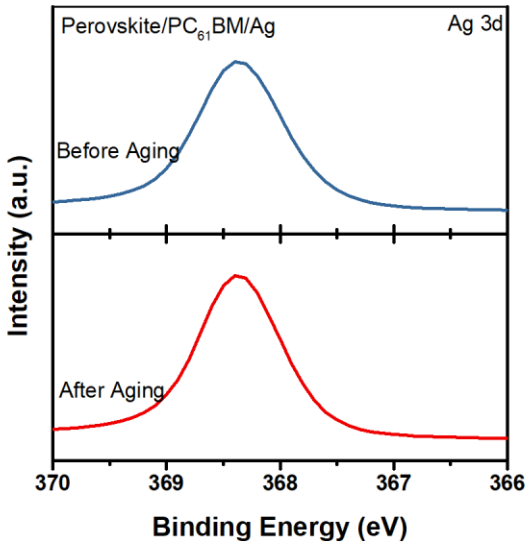

Figure S20. XPS spectra of Ag 3d obtained from the perovskite solar cell without TMA-PTC CIL before and after aging.

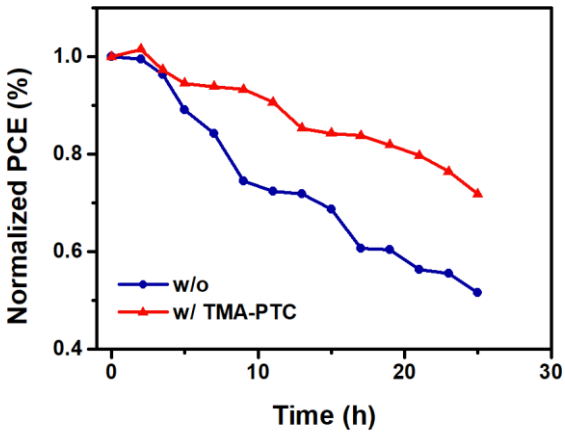

Figure S21. PCE decay of unencapsulated PeSCs with or without TMA−PTC as a function of exposure time in the air with a high relative humidity (>60%).

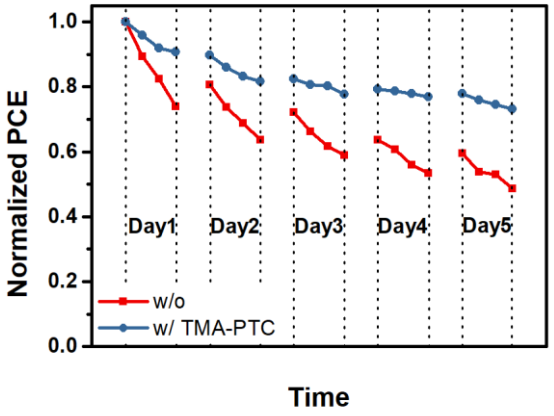

Figure S22. PCE decay of perovskite solar cell with and without TMA-PTC under the condition of light (1 sun illumination)-dark cyclings as a function of storage time.

Space charge limited current (SCLC) measurements

The electron-only devices were fabricated with a configuration of a configuration of ITO/$PC_{61}BM$/Perovskite/$PC_{61}BM$/TMA−PTC/Ag. The thickness of perovskite layer is ca. 310 nm, while the thickness of $PC_{61}BM$ layer is ca. 40 nm. The SCLC can be written as $J_{SCLC}=9\varepsilon_0\varepsilon_r\mu_e(V-V_{BI})^2/(8L^3)$: where J is current density, ε_r is dielectric constant of the perovskite materials (normally a value of 28.8 is used),[1] ε_0 ($8.8541878176\times10^{-12}$ F/m) is the permittivity of vacuum, μ_e is electron carrier mobility, L is film thickness, $V=V_{appl}-V_{bi}$, V_{appl} is the applied potential, and V_{bi} is the built-in potential resulting from work function difference between two electrodes (in this device, V_{bi}=0 V). To calculate the μ_e value, the log J-V line obtained in dark was fitted using a function of $y=a+2x$ and electron mobility (μ_e) was calculated from the y-intercept of the fitting line.

Reference.

[1] Yang, D.; Yang, R.; Ren, X.; Zhu, X.; Yang, Z.; Li, C.; Liu, S. Hysteresis−Suppressed High−Efficiency Flexible Perovskite Solar Cells Using Solid−State Ionic−Liquids for Effective Electron Transport. *Adv. Mater.* **2016,** *28*, 5206−5213.