Covalent Triazine Reducing the Defects by Coordination Roles and Inhibiting I[−] Migration by Anion-π Interaction for Efficient Perovskite Solar Cell

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Fig. S1. Synthesis of crystalline CTFs.



Fig. S2. (a and b) TEM image of CTFs. (c and d) TEM mapping of C and N elements in CTFs.



Fig. S3. The AFM images of exfoliated nanosheets from the layered bulk CTFs.



Fig. S4. (a) N₂ adsorption (closed circles) and desorption (open circles) isotherms (77 K) of CTFs; (b) Pore size distribution curves of CTFs.



Fig. S5. The Tauc plot of the perovskite films.



Fig. S6. The optimized configuration of CTF+perovskite, in which the perovskite with different terminal surface: (a) perfect Pb-I surface, (b) Pb-I surface containing V_I , (c) perfect FA-I surface and (d) FA-I surface containing V_I .



Fig. S7. Common interpretation of coloring method of mapped function $sign(\lambda_2)\rho$ in IGMH maps.



Fig. S8. The SEM images of perovskite films with different concentration of CTFs modification (a) 0.00 mg/mL, (b) 0.17 mg/mL, (c) 0.20 mg/mL, and (d) 0.23 mg/mL.



Fig. S9. Grain size analysis of perovskite films with different concentration of CTFs modification (a) 0.00 mg/mL, (b) 0.17 mg/mL, (c) 0.20 mg/mL and (d) 0.23 mg/mL.



Fig. S10. PL spectra of the perovskite films modified with different concentrations of CTFs.



Fig. S11. *J*–*V* curves of devices modified with different concentrations of CTFs.



Fig. S12. Box charts of (a) PCE, (b) J_{SC} , (c) V_{OC} and (d) FF of the PSCs.



Fig. S13 Box charts of (a) *PCE*, (b) J_{SC} , (c) V_{OC} and (d) *FF* of PSCs with different modification.



Fig. S14. Statistical histogram of *PCEs* of corresponding devices.



Fig. S15. Steady-state current density and *PCE* of the PSCs for 600 s measured at the maximum power point.



Fig. S16. UV–vis absorption spectra of perovskite films illuminated under simulated solar light (AM1.5G, 100 mW cm⁻²) for 180 min.



Fig. S17 Thermal stability of the unpackaged devices by heating them at 60 ± 5 °C in an Ar-filled glove box.



Fig. S18. Stability tracking of unpackaged devices stored in moisture-proof boxes (1-2%RH).



Fig. S19. Stability tracking of unpackaged devices stored in air (30±5%RH).



Fig. S20.Time-dependent stability measurements under continuous light illumination.



Fig. S21. PCE Evolution of PSCs under MPP tracking and continuous illumination.



Fig. S22. Stability of the device in an atmosphere filled with oxygen.



Fig. S23. XRD patterns of (a) control perovskite film and (b) target perovskite film aging in air condition with the humidity of $60\pm5\%$ RH.



Fig. S24. Diffraction peaks intensity ratio of PbI_2 and (110) plane of perovskite.



Fig. S25. Determination of lead concentration in damaged devices by inductively coupled plasma mass spectrometry (ICP-MS) after dripping test.

| Sample | $\tau_1/(ns)$ | A ₁ /(%) | $\tau_2/(ns)$ | A ₂ /(%) | $\tau_{ave}/(ns)$ |
|---------|---------------|---------------------|---------------|---------------------|-------------------|
| Control | 19.32 | 0.805 | 44.71 | 0.195 | 28.44 |
| Target | 21.33 | 0.632 | 55.99 | 0.368 | 42.28 |

 Table S1. TRPL data of perovskite films.

 Table S2. Energy level parameters of materials used in the device.

| Sample | E _{cut-off} (eV) | $E_{F, edge}$ (eV) | E _{VB} (eV) | Eg (eV) | E _{CB} (eV) |
|---------|------------------------------|--------------------|-------------------------|------------|-------------------------|
| Control | 16.84 | 1.08 | -5.44 | 1.53 | -3.91 |
| Target | 16.98 | 1.18 | -5.40 | 1.53 | -3.87 |

| Device | Scan | $J_{\rm sc}$ (mA/cm ²) | V _{oc} (V) | FF (%) | PCE (%) | HI |
|-------------------|---------|------------------------------------|------------------------|-----------|------------|-------|
| Control Target | Forward | 24.84 | 1.09 | 78.52 | 21.26 | 0.034 |
| | Reverse | 24.43 | 1.09 | 77.11 | 20.53 | |
| | Forward | 25.21 | 1.12 | 81.62 | 23.05 | 0.024 |
| | Reverse | 25.16 | 1.12 | 79.81 | 22.49 | |

Table S3. Photovoltaic performances parameters of the devices measured in reverse and forward scan under AM 1.5 G 1 sun illumination of 100 mW/cm².

 Table S4.
 Summary of the photovoltaic parameters of the PSCs with different concentrations of CTFs additive.

| CTFs | $J_{ m sc}$ | $V_{ m oc}$ | FF | PCE |
|---------|-----------------------|-------------|-------|-------|
| (mg/mL) | (mA/cm ²) | (V) | (%) | (%) |
| 0.00 | 24.84 | 1.09 | 78.52 | 21.26 |
| 0.17 | 24.96 | 1.12 | 79.26 | 22.16 |
| 0.20 | 25.21 | 1.12 | 81.62 | 23.05 |
| 0.23 | 25.13 | 1.12 | 79.30 | 22.32 |

| Device | $J_{\rm SC}$ (mA/cm ²) | V _{OC} (V) | FF (%) | РСЕ (%) |
|---------|------------------------------------|------------------------|------------|------------|
| Control | 24.62±0.59 | 1.09±0.04 | 75.46±3.46 | 20.66±1.22 |
| Target | 24.98±0.45 | 1.11±0.03 | 78.81±3.07 | 22.20±1.20 |

 Table S5. Averaged performances parameters of the 20 devices.