

High Efficiency Inorganic Perovskite Solar Cells Based On Low Trap Density and High Carrier Mobility CsPbI₃ Films

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Inorganic perovskite CsPbI₃ has exhibited promising performance in single-junction solar cells, but the grain boundaries (GBs) in its film cause the formation of the defects with deep energy levels (such as iodide vacancy (V_I)) and impede the transport of carriers, worsening the efficiency and stability of the solar cells. Here, a CsPbI₃ precursor is devised with thiophenol series ligands (TP-ligands) containing both S—H and π -conjugated molecules. The strong interaction between the S—H group (Lewis base) and PbI₂ (Lewis acid) suppresses the formation of PbI₄²⁻ in perovskite solution, thus suppressing the formation of V_I in the film accordingly. The terminal groups, such as -F and -NH₂, are employed to achieve an appropriate evaporation speed of the ligands and prevent the oxidation of the thiophenol group, then a high-quality perovskite film with low trap density is obtained. In addition, the functional group π -conjugated molecules provide additional carrier transmission channels at the GBs to increase carrier mobility, which facilitates the exciton separation and prolongs the charge lifetime. The CsPbI₃ solar cell shows a considerable power conversion efficiency of 20.1% with an open-current voltage of 1.18 V and a high fill factor of 83.5% and excellent working stability.

Several attempts have been made to stabilize the CsPbI₃ black phase. For instance, Br⁻ was introduced as the substitution for I⁻, increasing the τ value.^[5–7] Unfortunately, the light absorption range becomes narrower correspondingly with the Br component increasing. Dimensionality engineering, such as 2D perovskites and zero-dimensional perovskite quantum dots, is another option.^[8–13] The high surface energy provides a strain that prevents the [PbI₆]⁴⁻ octahedron from twisting. However, this process generates dozens of grain boundaries (GB) in the film, which may cause a large amount of energy loss in the carriers, leading to a shorter carrier diffusion length. Similar to dimensionality engineering, organic capping also can offer a surface strain to inhibit the phase transformation.^[14,15] What's more, the hydrophobicity of organic materials may lessen moisture erosion. Moisture exposure may trigger the phase transition by lowering

the phase transition barrier.^[16] Nonetheless, owing to their poor electrical conductivity, organic molecules will obstruct carrier transmission, resulting in the poor power conversion efficiency (PCE) of the devices. Nowadays, the most popular technique for CsPbI₃ fabrication is dimethylammonium iodide (DMAI) doping, resulting in a β -phase perovskite film with high efficiency and phase stability.^[17–19] However, because of the evaporation of DMAI during the annealing stage, it still suffers from poor morphology and vacancy defects.^[20–23]

In addition, the sol-gel technique is commonly used for the fabrication of the perovskite film, leading to a polycrystalline absorber layer. Defects would unavoidably arise in the GBs of the films during the production process. Among these defects, iodide vacancy (V_I) is commonly regarded as a type of defect with deep energy levels that acts as Shockley-Read-Hall nonradiative recombination centers, serving as photoluminescence quenching sites and thus resulting in the loss of photogenerated carriers.^[24,25] The phase transformation of the CsPbI₃ perovskite can likewise be easily induced by these defects. According to the Lewis acid/base theory, the ligands with (L) and bidentate (LL) groups usually act as the Lewis base and have the effect of V_I passivation, such as O-donor, S-donor, and N-donor.^[26] Consequently, the Lewis base S—H has a chance to form an adduct by interacting with the PbI₂, which is known to be a Lewis acid, thus passivating the V_I defect.^[27] In addition, the π -conjugated molecules normally process superior electrical conductivity because of the unique natures of coplanar π -electrons and the

1. Introduction

Cesium lead halide perovskite (CsPbI₃) has received a lot of attention for photovoltaics in recent years because the A-site organic cation is replaced by cesium to overcome the intrinsic instability of organic-inorganic hybrid perovskite.^[1–3] With a bandgap of 1.68 eV, the β -phase CsPbI₃ perovskite is a viable choice for both high-efficiency single-junction perovskite solar cells (PSCs) and top cells with a wide bandgap in tandem solar cells. The toughest issue of CsPbI₃ perovskite is the nature of structural instability. The size-unmatchable Cs ions barely hold up the construction of [PbI₆]⁴⁻ octahedron, leading to the transformation from black phase (α phase, β phase, γ phase) to yellow phase (no-perovskite phase).^[4]

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existence of intermolecular π - π interaction.^[28] It could be speculated that the functional group would improve the ability to transfer carriers through the perovskite interfaces and GBs.

Here, we devise thiophenol series ligands (TP-ligands) as the multifunctional molecular modulators in the perovskite precursor solution, which contains both the S-H and π -conjugated molecules. The terminal groups such as -F and -NH₂ are employed to adjust the dipole moments of thiophenol to prevent it from oxidation. TP-ligands not only modify the halide vacancy, but also provide additional beneficial channels for carrier extraction at the GBs of the perovskite film. Moreover, the crystallization of CsPbI₃ films is improved, leading to the removal of pinholes in perovskite film. As a result, an impressive PCE of 20.1% was achieved with an open-current voltage (V_{OC}) of 1.18 V and a high fill factor (FF) of 83.5%. Furthermore, the thiophenol-containing compounds have contributed exceptional long-term durability to perovskite films because of their hydrophobicity.^[29] The CsPbI₃ devices with thiophenol modification exhibit excellent performance in both aging and working stabilities at the maximum power point (MPP).

2. Results and Discussion

We devised three TP-ligands with different terminal groups: 4-Fluorothiophenol (4-FTP), 4-aminothiophenol (4-ATP), and 2-aminothiophenol (2-ATP). We first studied the behavior of the TP-ligands in the CsPbI₃ precursor solution. The perovskite precursor was diluted and measured by ultraviolet-visible

(UV-vis) spectroscopy to explore the effect of TP-ligand on the colloidal characteristics, as shown in Figure 1a,b, and Figure S1 (Supporting Information). When the addition concentration of the precursor was lower than 120 μ M, there was an obvious absorption peak of PbI₂ at 325 nm. A red shift occurred gradually in the UV-vis spectra as the precursor concentration increased. In addition, new peaks at 375 and 425 nm arose and grew, meaning that PbI₂ underwent complexation to produce soluble PbI₃⁻ and PbI₄²⁻ (the coordinated plumbate species).^[30,31] We investigated the effects of the TP-ligands on the dynamics of PbI₃⁻ and PbI₄²⁻ production. Figure 1c demonstrates the increased rate of the peaks at 375 and 425 nm with the increase of solution concentration, representing the increased amount of PbI₃⁻ and PbI₄²⁻. The lower growth rate of absorption of PbI₃⁻ and PbI₄²⁻ in the TP-ligand CsPbI₃ solution illustrates that the coordination between TP-ligands and PbI₂ suppresses the formation of PbI₄²⁻. Previous works have demonstrated the formation of PbI₄²⁻ in the precursor would increase the defect density in the product perovskite film.^[32] Thus, the introduction of TP-ligands will help to prepare low defect density perovskite films.

The density functional theory (DFT) calculations were carried out to further demonstrate the behavior of the TP-ligand (4-ATP). The results show the adsorption energy (ΔE) between the S-H group and uncoordinated Pb²⁺ on the CsPbI₃ film in a value of -0.326 eV, indicating that it is easy for TP-ligands to be adsorbed at iodide vacancies (V_I) on the surface of CsPbI₃ during the self-assembly process, as illustrated in Figure S2 and Table S1. (Supporting Information) To check for the reliability of

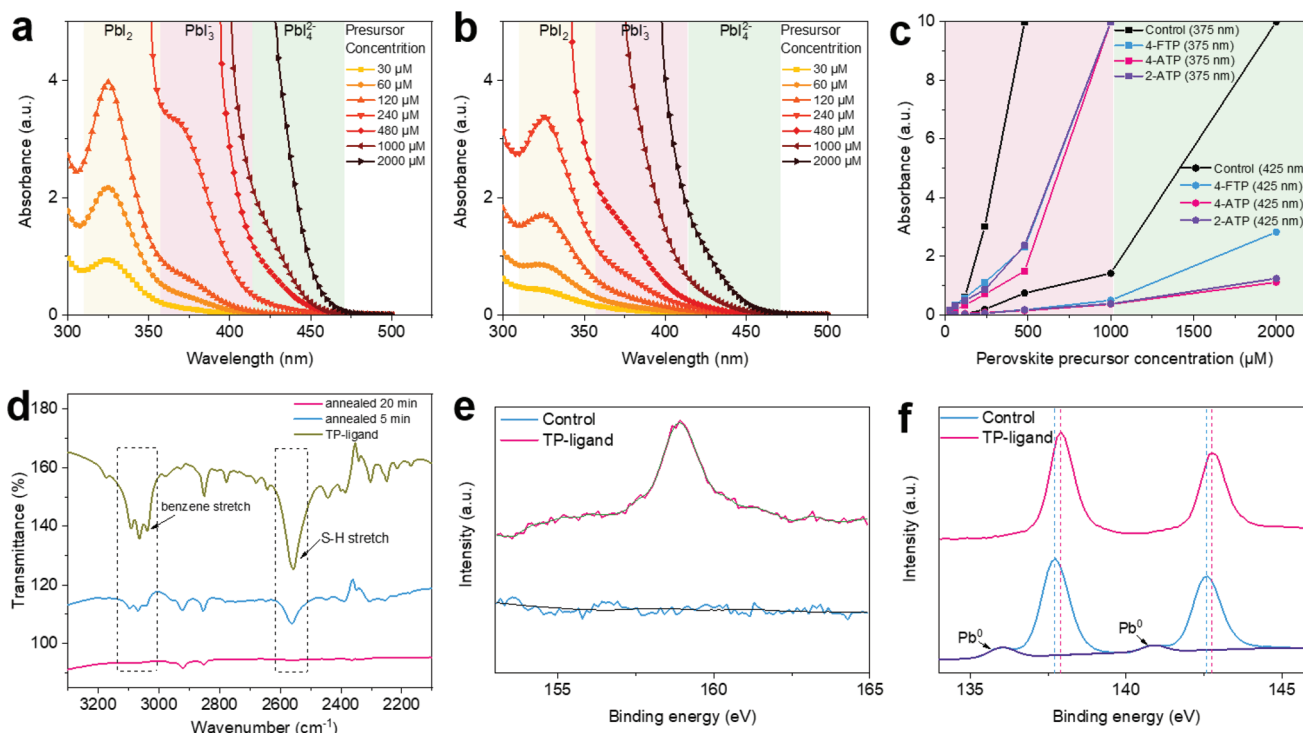


Figure 1. The UV-vis to track the different concentrations of a) control and b) 4-ATP CsPbI₃ precursor. The yellow region, pink region, and green region illustrate the absorption peak range of PbI₂, PbI₃⁻ and PbI₄²⁻, respectively. c) The increased rate of the peaks at 375 and 425 nm of the perovskite solution. d) FTIR spectra of the TP-ligands, TP-ligand CsPbI₃ films annealed 5 and 20 min. XPS data for e) S 2p and f) Pb 4f_{7/2} and Pb 4f_{5/2} core-level spectra in control and TP-ligand CsPbI₃ films.

these results, the energy dispersive spectroscopy (EDS) of the TP-ligand CsPbI₃ film is presented in Figure S3 (Supporting Information). The S elements were homogeneously distributed across the perovskite film. The molecular interactions of the ligand with perovskite film were examined by Fourier transform infrared spectroscopy (FTIR), as shown in Figure 1d. By zooming in the spectra, several weak peaks were observed at 1560 and 3100 cm⁻¹ from the TP-ligand CsPbI₃ film (20 min of annealing), which are the vibration peaks of S–H and benzene (Figure S4, Supporting Information), respectively. The peak characteristic of the S–H stretching vibration was found in the FTIR spectra at 1555 cm⁻¹ for the TP-ligand solution and 1560 cm⁻¹ for perovskite film. The S–H stretching underwent broadening and shifted to a longer wavelength, indicating the interaction of S–H with uncoordinated Pb²⁺. To confirm this result, the perovskite film was further studied by X-ray photoelectron spectroscopy (XPS), as shown in Figure S5a, (Supporting Information). The XPS spectra of the TP-ligand CsPbI₃ and control film in the S 2p level range are shown in Figure 1e, with the S 2p peak visible in the TP-ligand CsPbI₃ spectrum. Figure 1f displays the Pb 4f core spectra of the TP-ligand CsPbI₃ and control films. The control film shows two small peaks located at 136 and 140.8 eV, which are caused by the presence of metallic Pb (Pb⁰), a primary deep defect state that substantially impairs the performance and long-term durability of perovskite optoelectronic devices.^[33] While the metallic Pb peaks disappeared in the TP-ligand CsPbI₃ film, suggesting the coordination between the ligands and Pb²⁺ ions responsible for the suppression of Pb⁰. In addition, the Pb 4f and I 3d peaks (Figure S5b, Supporting Information) in TP-ligand CsPbI₃ shifted to higher binding energy, indicating a decrease in electron density around these species due to the reduction of under-coordinated Pb²⁺ ions via interaction with S–H.

In our previous research, we found that the superfluous organic that accumulated at the GBs could obstruct charge transportation.^[34] If the residual organic without bounding to perovskite can be removed from the film during the annealing process, the efficiency of devices will be further improved. The volatility of TP-ligands was explored by thermogravimetric analysis (TGA), as shown in Figure S6 (Supporting Information). Under the annealing temperature, the 4-FTP, 4-ATP, and 2-ATP exhibit evaporation velocity from high to low orderly. X-ray diffraction (XRD) measurements were employed to trace the crystallization of the perovskite films, as shown in Figure S7 (Supporting Information). The stronger diffraction peaks originating from the β phase of CsPbI₃ reveal the high quality of the 4-ATP and 2-ATP CsPbI₃ films, which can be attributed to their slower evaporation velocity than that of 4-FTP. The slow evaporation velocity of the additive suppresses nucleation, leading to a decrease in the film growth, which is available for obtaining high-quality and morphology-improved perovskite films.^[35] As demonstrated in Figure 2a–d, the TP-ligands have a positive impact on the morphology of the perovskite film, where the cracks and pinholes in the original perovskite films appear to have been repaired. As shown in Figure S8 (Supporting Information), the denser morphology of the films results in a lower leakage current, leading to a higher FF for solar cells. In addition, the scanning electron microscopy (SEM) patterns show the bigger grains and fewer GB of the TP-ligand films than those

of the control sample, resulting in fewer nonradiative combinations due to low trap density. In Figure 2d, the white content should be 2-ATP, indicating the residual after the annealing process because of its low evaporation velocity at the annealing temperature. Figure S9 (Supporting Information) displays the macroscopical photograph of the films after the annealing process, showing the higher phase stability of TP-ligands. Based on the abovementioned results, we proposed the mechanism of TP-ligands for the formation of CsPbI₃ film: the complexation between TP-ligand (electron-pair donor) and PbI₂ (electron-pair acceptor) form a colloid complex in the perovskite precursor. The formation of PbI₄²⁻ is suppressed in perovskite solution, leading to a low trap density of charge recombination in the CsPbI₃ film. Furthermore, the morphology and crystallinity of the perovskite film are also improved. Whereafter, because of the evaporation effect, the extra TP-ligands can be eliminated during the annealing process, and the remnant would combine with the uncoordinated Pb²⁺ by the S–H bond, thus passivating the halide vacancies, as shown in Figure 2e.

To investigate the recombination kinetics of the 4-FTP, 4-ATP, and 2-ATP on the CsPbI₃ films, photophysical characteristics were performed. Figure 3a shows the time-resolved photoluminescence (TRPL) results of the TP-ligand and control films. The biexponential fitting parameters are presented in Table S2 (Supporting Information). The PL lifetime of the TP-ligand samples shows the longer lifetime of carriers and the smaller τ_1 ratio of TP-ligand CsPbI₃ suggests the effective suppression of the formation of nonradiative trap centers. Figure S10 (Supporting Information) displays the UV–vis and PL spectrum, with the same absorption edge of 736 nm consistent with the bandgap of β phase CsPbI₃. In addition, the stronger PL peak of the TP-ligand CsPbI₃ reveals less nonradiative recombination. PL intensity scanning images confirm this result, as shown in Figure 3b,c. In contrast to the control film, the TP-ligand CsPbI₃ exhibits a more uniform emission distribution and a higher absolute PL intensity, which is consistent with the average PL data in Figure S10 (Supporting Information). We utilized space-charge-limited-current (SCLC) methods to determine the defect density in the TP-ligand CsPbI₃ and control films. Dark current–voltage (*J*–*V*) characteristics are collected for single-carrier devices with an FTO/SnO₂/Perovskite/PCBM/Ag architecture, as shown in Figure 3d. The trap-filled limit voltage (*V*_{TFL}) values of the trap-filling regime are 0.212 V for the control film, 0.171 V for 4-FTP-CsPbI₃, 0.138 V for 4-ATP-CsPbI₃, and 0.199 V for 2-ATP-CsPbI₃ film, respectively. The trap density has effectively decreased from 9.51×10^{15} to 6.19×10^{15} cm⁻³ by 4-ATP ligand incorporation. Figure 3e depicts the relationship between *V*_{OC} and light intensity in the solar cells based on the TP-ligand CsPbI₃ and control films. The cross section SEM image described the device with a structure of FTO/SnO₂/CsPbI₃/spiro-OMeTAD/Ag, as shown in Figure 3g. The TP-ligand CsPbI₃ device shows a slope of 1.46 kT/*q* (4-FTP), 1.39 kT/*q* (4-ATP), and 1.36 kT/*q* (2-ATP), (where *k* is the Boltzmann constant, *T* is the absolute temperature, and *q* denotes the elementary charge), which is significantly lower than that of the control device (2.12 kT/*q*), indicating that the trap-assisted recombination is suppressed by the TP-ligands. In addition, the carrier extraction and recombination processes in the solar cells were carefully analyzed through electrochemical impedance

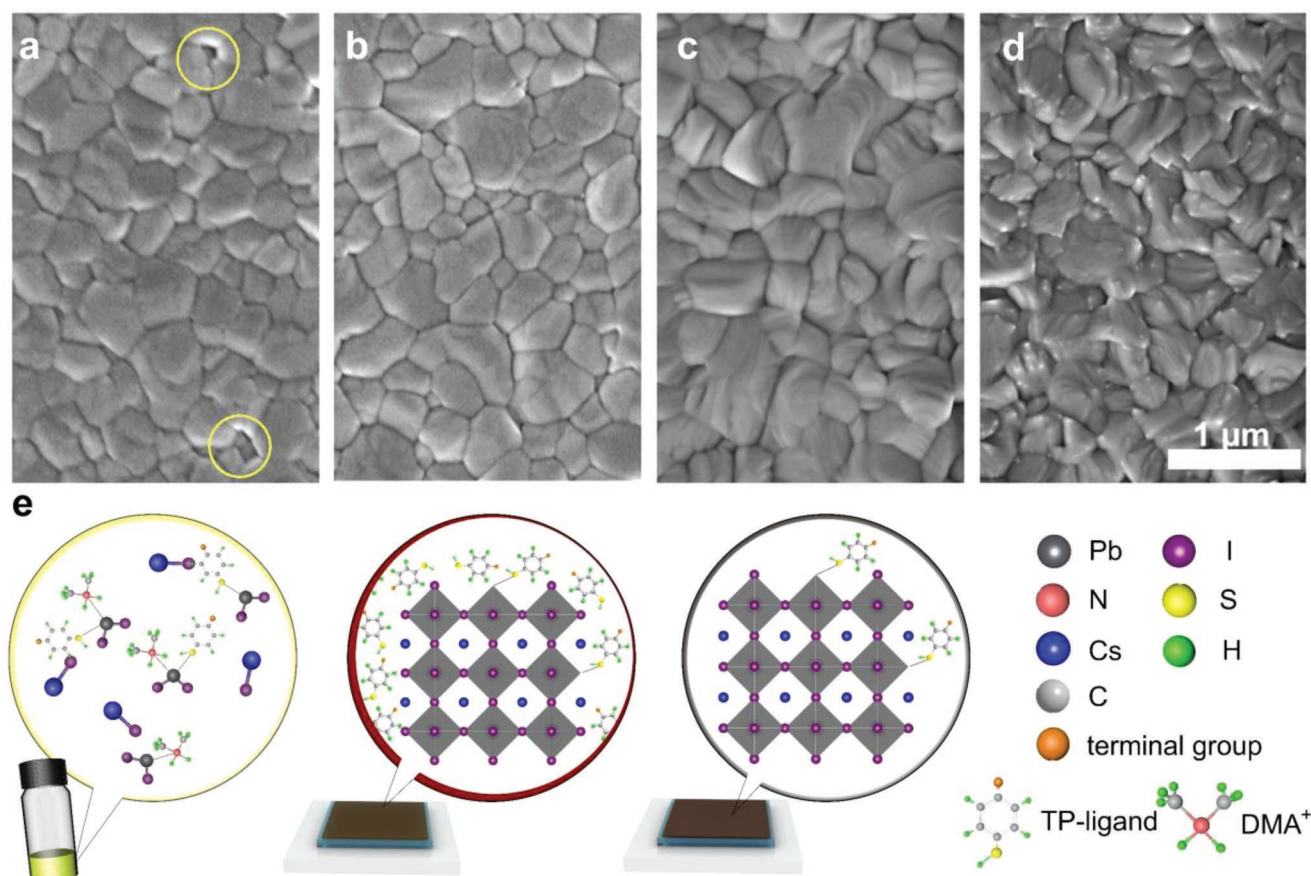


Figure 2. Top-view SEM images of the a) control film, b) 4-FTP CsPbI₃ film, c) 4-ATP CsPbI₃ film, and d) 2-ATP CsPbI₃ film. The pinholes are marked and the scale bar is 1 μm . e) Schematic diagram of the formation of CsPbI₃ films with introduction of TP-ligands.

spectroscopy (EIS) under illumination, as in Figure 3f. The fitting parameters are listed in Table S3 (Supporting Information). In the present planar PSCs, the smaller series resistance (R_s) and charge transfer impedance (R_{ct}) reveal the better interfacial contact of the TP-ligand CsPbI₃ device, leading to the suppressed recombination and effective charge extraction. Meanwhile, the largest impedance of the 4-ATP-CsPbI₃ device corresponds to a higher-quality active layer with fewer shunt routes due to fewer pinholes and fractures.

We prepared solar cells and their structure as shown in Figure 3g. Compared with the control CsPbI₃ device, the PCEs of the solar cells based on TP-ligand CsPbI₃ films are improved, as shown in Figure 3h and Table 1. The control CsPbI₃ device shows a typical PCE of 17.0% with a short-circuit current density (J_{sc}) of 20.1 mA cm^{-2} , a V_{oc} of 1.08 V, and FF of 78.2%. The 4-FTP and 2-ATP solar cells showed improved PCEs of 19.1% and 17.4%, respectively. Surprisingly, the optimized 4-ATP device delivered the best performance with a PCE of 20.1%, V_{oc} of 1.18 V, J_{sc} of 20.4 mA cm^{-2} , and FF of 83.5%, which is owed to its high-quality film with low trap density. Besides, negligible hysteresis under forward scan was obtained, as shown in Figure S11 (Supporting Information). A bias voltage of 1 V was applied to the polarity of the device to assure the reliability of the J - V measurements, resulting in a stable PCE of 19.1%, as shown in Figure S12 (Supporting Information). Meanwhile, Figure 3i

shows the external quantum efficiency (EQE) spectrum, which demonstrates that the integrated photocurrent (19.6, 19.7, 19.3, and 19.3 mA cm^{-2}) for the 4-FTP, 4-ATP, 2-ATP and control devices, respectively, which confirm the accuracy of the device efficiency characterizations. To verify the repeatability of the device performance, Figure S13 (Supporting Information) illustrates the statistical PCE and the detailed parameters of the devices, revealing the higher V_{oc} of the TP-ligand CsPbI₃ solar cells. The performance of the 2-ATP devices shows a very low FF. It can be attributed to the residual 2-ATP cluster on the perovskite film GBs, which blocks the transportation of carriers, leading to a higher series resistor.

To explore the mechanism of the TP-ligands in high-performance CsPbI₃ solar cells, we studied the contribution of π -conjugated molecules to the film conductive property. From SCLC measurement (Figure 3d), the electronic carrier mobility can be calculated by the Mott-Gurney law in the single-carrier devices. The electronic mobility of the 4-FTP, 4-ATP, 2-ATP, and control CsPbI₃ films are $18.3 \times \text{cm}^2$, $21.2 \times \text{cm}^2$, $0.3 \times \text{cm}^2$, and $4.5 \times \text{cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, respectively. The 4-ATP and 4-FTP samples show the highest electronic mobility. The residual organic in the 2-ATP film GBs may inhibit the charge transmission, leading to low carrier mobility. Local surface potential and film conduction through local J - V measurements were studied by conductive atomic force microscopy (c-AFM) analysis under a

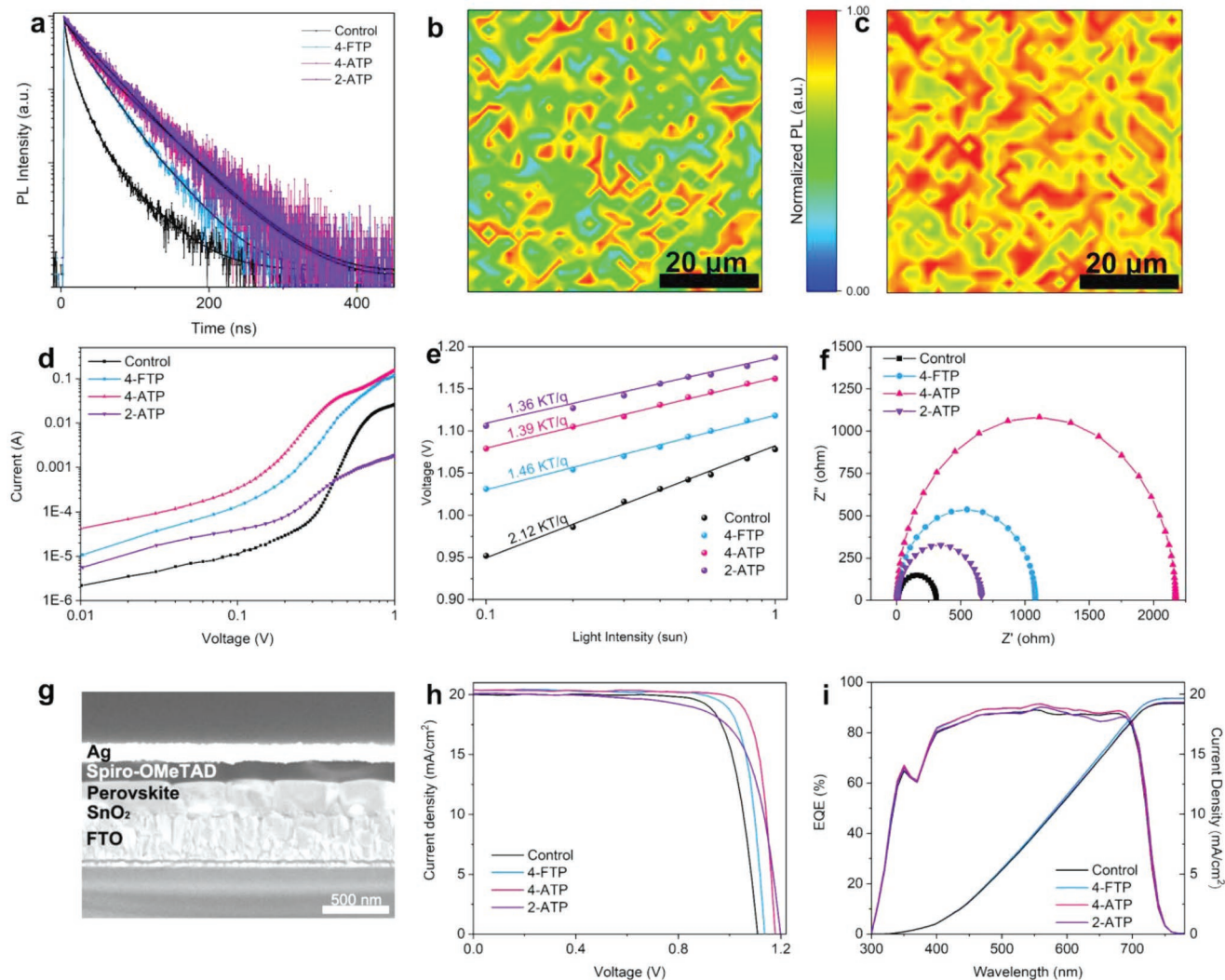


Figure 3. a) TRPL of CsPbI₃ films. PL intensity scanning images of b) controls and c) TP-ligand CsPbI₃ films and the scale bar is 20 μm . d) SCLC patterns of control and TP-ligand CsPbI₃ devices with an FTO/SnO₂/perovskite/PCBM/Ag structure (electron-only devices). e) V_{OC} dependence of the light intensity of the control and TP-ligand CsPbI₃ devices. f) EIS pattern of TP-ligand CsPbI₃ and control PSCs under an illuminated condition. g) Cross-sectional SEM image of the PSCs. h) Current J - V curves of the four PSCs under AM 1.5G irradiation (100 mW cm^{-2}). i) EQE and integrated current density spectra of the four PSCs.

bias voltage of 2 V, as shown in Figure 4a,b. The 4-ATP-CsPbI₃ gave considerably larger currents in the entire measured area than that of the control sample. According to the AFM patterns of this region (Figure S14, Supporting Information), we observed the obvious abrupt decreased conducting current at GBs in the control sample, while inconspicuous in the 4-ATP-

Table 1. Photovoltaic parameters of the champion device upon the control, 4-FTP CsPbI₃, 4-ATP CsPbI₃, and 2-ATP CsPbI₃ under AM 1.5G, 100 mW cm^{-2} illumination.

Samples	V_{OC} [V]	J_{SC} [mA cm^{-2}]	FF [%]	PCE [%]
Control	1.08	20.1	78.2	17.0
4-FTP	1.14	20.4	81.9	19.1
4-ATP	1.18	20.4	83.5	20.1
2-ATP	1.20	20.1	72.0	17.4

CsPbI₃, elucidating the additional conductivity channel for carrier extraction at GBs passivated by TP-ligands.

The Exciton behavior in the film was further investigated by normalized steady-state temperature-dependent PL and the spectra were mapped in color-coded images, as shown in Figure 4c,d. The strong peaks appeared both in the control sample and 4-ATP-CsPbI₃ at 100 K. As temperature further increases, PL intensity decreases correspondingly, while the 4-ATP-CsPbI₃ shows a larger decline than the control sample, as shown in Figure S15 (Supporting Information). To understand the reason, the exciton binding energy (E_b) of the two films can be extracted, which represents the overall energy barrier for excitons to be bound to nonradiative centers. As shown in Figure S16 (Supporting Information), the control film shows the E_b of 13.9 meV while it is down to 7.5 meV for the 4-ATP-CsPbI₃ film, which is beneficial to the separation of the exciton, giving rise to more free carriers.

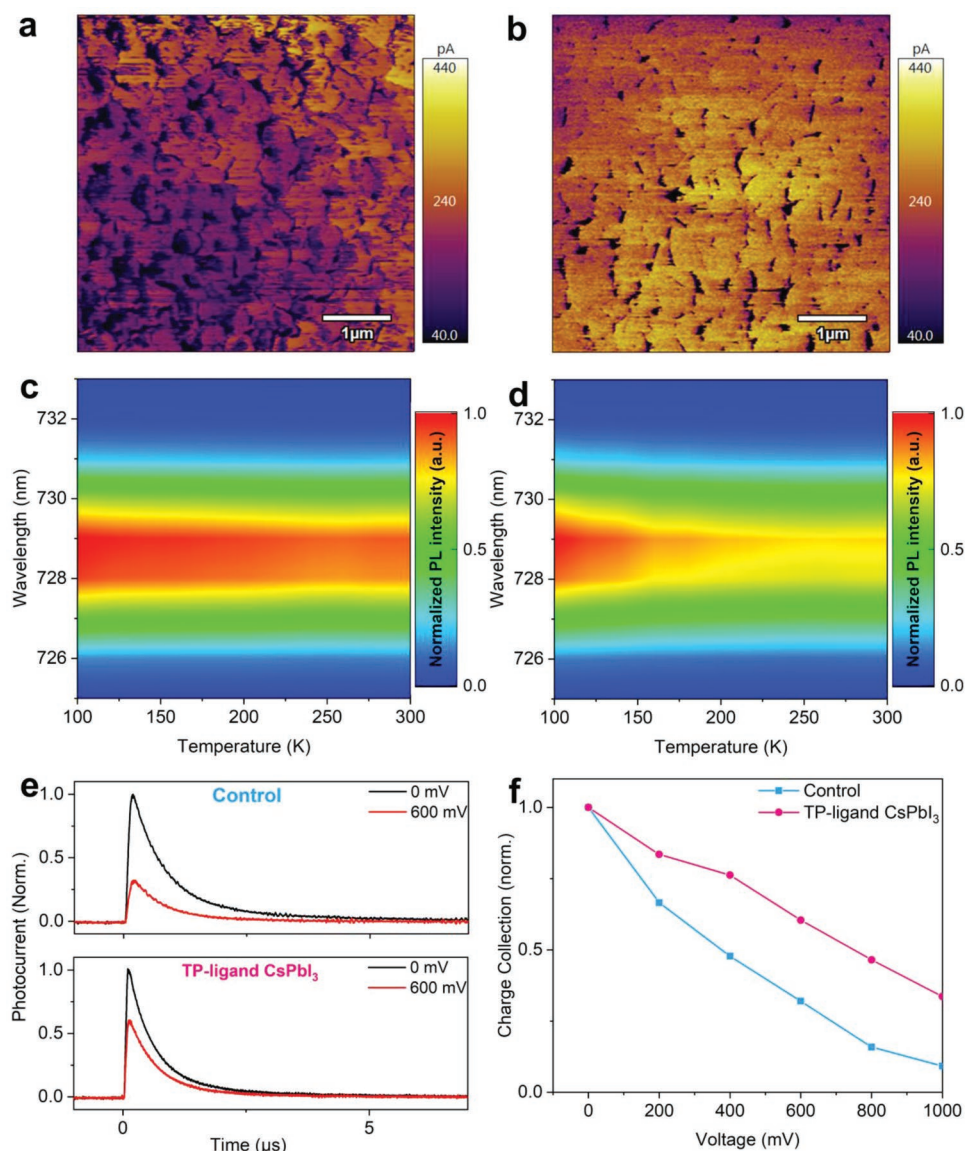


Figure 4. c-AFM images of a) control film and b) 4-ATP CsPbI₃ film. Temperature-dependent PL of c) control film and d) 4-ATP CsPbI₃ film. e) TPC results of the cells at bias voltages of 0 and 600 mV. f) Normalized charge collection of the cells calculated with $\Delta Q/\Delta Q$ (200 mV), where ΔQ is the total collected photo-induced charge by integrating the TPCs.

Aside from charge recombination, an increased film conductivity may have an impact on the carrier transport capabilities and final charge collection of devices.^[36–38] The transient photocurrent (TPC) was measured at various bias voltages as shown in Figure 4e. The energy of the incident pulse laser remains unchanged during the measurement, as the bias voltage increases, the charge collection efficiency declines. When the bias voltages are enhanced from 0 to 600 mV, the intensity of the photocurrent peak on the control sample decline to 30%, whereas the TP-ligand CsPbI₃ device still retains 60%. The total charge collection can be calculated by integrating the photocurrents. Hence, the total collected charge of the different solar cells at varying bias voltages is finally computed and summarized in Figure 4f. At high bias voltages, the charge collection

of the solar cells decreases because of the increase in charge recombination. However, a lower descent rate suggests faster charge transmission, which reduces carrier loss and increases charge collection efficiency, leading to a higher FF of the device. TPC and transient photovoltage (TPV) measurements of the various devices were also performed at a bias voltage of 200 mV. As shown in Figure S17 (Supporting Information) (fitted data in Tables S4 and S5, Supporting Information). In the TPC pattern, the TP-ligand CsPbI₃ device obtained a shorter decay time of 0.72 μs, indicating a faster charge transport and extraction process than the control sample, which has a decay time of 1.73 μs. The TP-ligand CsPbI₃ device has a longer electron lifetime of 4.76 ms based on the voltage decay time for each device. The longer photovoltage decay time of the TP-ligand CsPbI₃ device

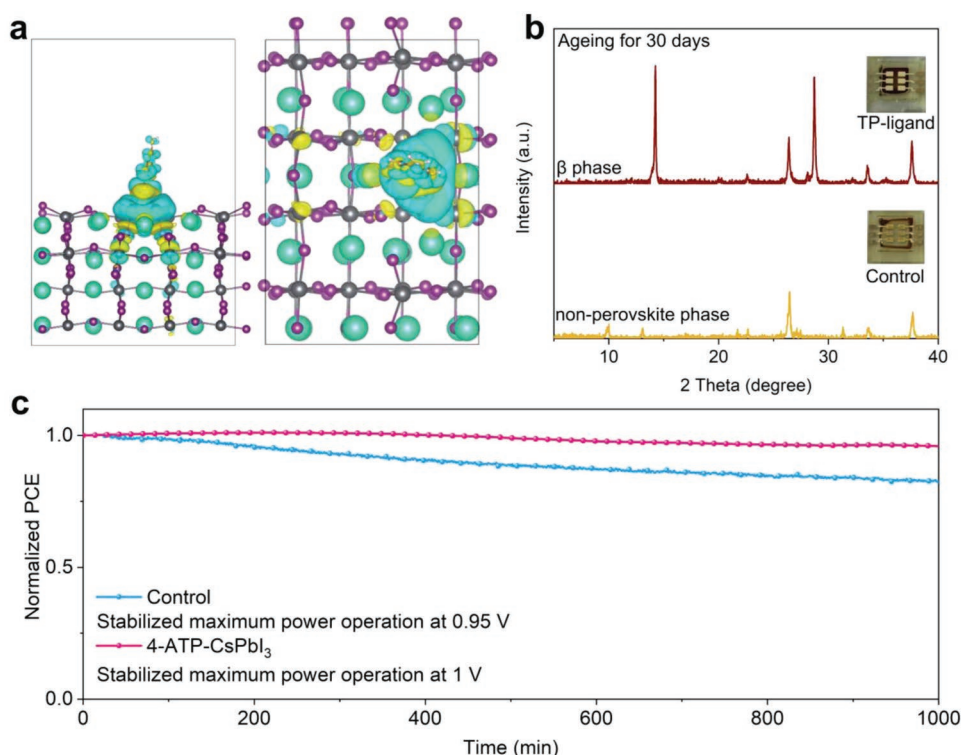


Figure 5. a) Bader charge analysis of the V_I defect passivated by TP-ligands. b) Photographs and XRD patterns of various CsPbI₃ films aged for 30 days. c) The operational stability of the perovskite devices at their MPP for 1000min.

is likely due to the preferred orientation, which allows for better carrier flow between perovskites and carrier transport layers.

To provide a complete theory between the TP-ligands and carrier transfer, we further estimated the electron transfer nature and electronic interactions with Bader charge analysis. As shown in **Figure 5a**, there is an increased charge transfer of 0.094 e⁻ from the I atoms of the CsPbI₃ structure to the TP-ligands. In addition, charge transfer is mainly observed between the π -conjugated molecule and the I atoms. We encapsulated the devices in an environment with $\approx 20\%$ relative humidity and tested their long-term stability. Figure S18 (Supporting Information) shows that after one month of storage, the TP-ligand CsPbI₃ devices maintained 83% of their initial PCE and remained in the black phase, whereas the control sample lost efficacy and the color turned yellow. XRD pattern revealed that the TP-ligand CsPbI₃ film was still in the β phase while the control film had converted to the non-perovskite phase, as shown in **Figure 5b**. The operational stability was also measured, as shown in **Figure 5c**. The TP-ligand-CsPbI₃ device showed negligible attenuation following 1000 min of continuous AM1.5 at their MPP. As a contrast, the control device lost nearly 20% efficiency at 1000 min. The elevated stabilities are attributed to the hydrophobicity of TP-ligands. To prove it, **Figure S19** (Supporting Information) displays the water contact angle measurement, which can verify the improvement of hydrophobicity for perovskite films. The water contact angle increases from 66° to 80° after TP-ligand passivation, leading to the hydrophobic surface chemistry of TP-ligand CsPbI₃ films.

3. Conclusion

We introduced multifunctional TP-ligands into perovskite precursors to obtain a stable CsPbI₃ film with low trap density and high carrier mobility, resulting in efficient all-inorganic PSCs. The TP-ligands coordinated with Pb²⁺ ions by the S–H group in the precursor, thus suppressing the formation of PbI₄²⁻ to reduce the defect density in the product perovskite film. In addition, the coordination of TP-ligands to the Pb–I octahedrons improved the morphology and crystallinity of the CsPbI₃ film, considerably reducing the nonradiative charge recombination center. The extra TP-ligands with an evaporation effect can be removed during the annealing process, while the ones with low evaporation velocity (such as 2-ATP) will accumulate at the GBs and on the surface, thus hindering the carrier transport. The functional group of π -conjugated molecules process accelerated the charge separation because of the unique features of coplanar π -electrons. As a result, a progressive PCE of 20.1% was achieved for 4-ATP CsPbI₃ PSC with a high V_{OC} of 1.18 V and strikingly FF of 83.5%. Besides, the PSC exhibited excellent operational stability due to the hydrophobic nature of TP-ligands. These results point in a new direction to fabricate efficient and stable all-inorganic PSCs.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

efficiency, inorganic metal halide perovskites, passivation, solar cells, thiophenol ligands

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