

Bifunctional Multidentate Linker Tailoring Interfacial Dipole and Residual Stress to Enhance Photovoltaic Performance

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Abstract

Buried layer crystallization at an electron transport layer interface is seldom discussed but crucial for obtaining a high-quality perovskite layer. This study demonstrates a novel interfacial engineering concept by using 2-aminoethylphosphonic acid (APA) to bridge the mechanical and electronic disparities at the SnO_2 /perovskite interface. The bifunctional synergy of the APA molecule, which serves as both an electronic dipole and a mechanical geometrical tuner. The flexible backbone of APA renders it to alleviate the strain by over 20% through releasing residual compressive stress. The electron-withdrawing phosphonate and electron-donating amino headgroups anchor preferred-orientationally onto SnO_2 and perovskite terminal facets. The directional dipole and releasing stress help expand the quasi-Fermi level splitting and accelerate electron transfer from the perovskite layer to the electron transport layer of SnO_2 . The champion device can achieve power conversion efficiency of 22.92% with over 1,500 h stability in air without encapsulation. By harmonizing molecular orientation with mechanical stress relief, this study establishes a new paradigm for high-performance and durable perovskite photovoltaics.

Introduction

Organometal halide perovskite solar cell (OHPSC) has advanced rapidly over the past decade.[1–4] The corresponding photovoltaic performance stands out from the other types of photovoltaics owing to its ambipolar transport, long carrier diffusion length, and high visible light absorption coefficient.[5–7] The above properties confer organo-metal halide perovskite (OHPVK) materials to narrows the photovoltaic gap between single crystal and polycrystal condition with appreciable defect density. Although OHPVKs are considered as high defect tolerance photovoltaic materials, their trap states in a band structure still perturb the minority carrier transport.[8] That deteriorates the reliability and causes hysteresis when they are under long-term operation.[9, 10]

In the next-phase progressing to approach the ceiling of photovoltaic performance and long-term reliability of OHPSCs, many efforts focus on defect passivation to alleviate the defect induced trap states.[11–13] In addition to defects in OHPSCs, the lattice stress in OHPVK materials recently catches lots of attention.[14–17] The containing stress is believed to deform the lattice of OHPVKs and results in both crystal structure instability and reduce of quasi-Fermi-level splitting (QFLS) in an OHPSC.[18] That limits the attainable long-term stability and open-circuit voltage (V_{OC}) loss of a device. Accordingly, strain engineering in OHPVKs offers a route to approach the ceiling of OHPSCs by enlarging degree of QFLS and increase a V_{OC} of OHPSCs. Surface strain manipulation is reported to effectively lower bimolecular recombination and to reduce V_{OC} loss of OHPSCs by modulating charge-emitting states. The corresponding devices minimize V_{OC} loss to 307 mV, which is the record in p-i-n OHPSCs.[19] In parallel, a facet-complementary strategy with formamidine-based additive, FPPC, demonstrates the relationship between strain control and α -FAPbI₃ structural robustness. The strong coordination affinity of FPPC regulates the facet orientation, optimizes strain accumulation of OHPVK films, and improves performance and stability of OHPSCs.[20] Nanoscale mapping reveals pronounced intra- and inter-grain strain variations and uncovers evidence for dislocation-induced antiphase boundary formation.[21, 22] The local strain heterogeneity and extended defects, dislocations, markedly affect the performance. It is believed to be one of critical reasons for instable Sn-based OHPSCs.[23] Spatial strains causes random crystal stacking and inducing shallow- and deep-level trap states in OHPVK films. By composition optimization, the spatial strains can effectively release and improves non-radiative recombination. Based on the improvement, the voltage deficit reduces from 0.148 to 0.114 V.[24] Long-ligand assist strategy has also reported to fragment 3D OHPVK structure into 2D slabs. That alleviates structural bonding between corner-sharing PbI₆ octahedra and relaxing the in-plane tensile strain.[25]

The studies for strain relaxation of an OHPVK mainly focus on the manipulation through additive or tuning corner-sharing PbI₆ octahedron during precursor state or the film formation stage.[26, 27] Few studies gain insight the lattice mismatch caused between a beneath carrier transport layer and an OHPVK layer in the beginning stage of crystallization. The lattice misfit makes the intimate layer of OHPVKs grow under compressed/tensile stress condition. As the deposited film get thicker, the OHPVK layer inherits the strain and release the strain through destruction of ordered atom arrangement. In macroscope point of view, a buried layer is observed between a carrier transport layer and an OHPVK layer. Such a layer, namely dead layer, adversely influence charge transfer between a carrier transport layer and an OHPVK layer. The same conditions have already been observed in both n-i-p and p-i-n device structure. [28, 29] Therefore, optimizing the lattice misfit between carrier transport layers and OHPVK layers is promoting to resolve lattice strain in the beginning of crystallization.

Herein, 2-aminoethylphosphonic acid (APA) acts as a multidentate-dipole interfacial linker (MIL) that completes interfacial coordination and aligns energy levels at the SnO₂/OHPVK contact. The phosphate headgroup chemisorbs on SnO₂ (P-O-Sn), while the protonated amine bonds the OHPVK surface (coordination to under-coordinated Pb²⁺/H-bonding with I⁻), forming a self-aligned interphase with a net dipole. That favors electron collection by SnO₂. This dual anchoring relaxes residual lattice strain through distributing interfacial lattice misfit, suppressing lattice distortion, and accelerating electron transfer from the OHPVK to SnO₂. Both effects promote the photovoltaic performance from 20.32% to 22.92% of OHPSC with APA modified SnO₂ with 300 mV of voltage improvement.

Results and Discussion

Herein, we first focused on the interaction of MIL between electron transport layer (ETL) of SnO₂ surface and perovskite surface. Four ending functional groups including thiol (-SH), amino (-NH₂), carboxyl (-COOH), and phosphate (-PO₄), were selected to design a proper passivator to manage both dangling bond from SnO₂ and ionic defects at perovskite interface. Beyond defect passivation, the intermolecular dipole moment of these MILs is another decisive point to select the MIL.[30] A well-aligned interfacial dipole facilitates electron injection from the perovskite layer to the ETL by minimizing the interfacial potential drop and reducing energy loss during charge transportation.[31] As a result, we selected two specific categories as ending functional groups on the molecules as shown in Fig. 1 (a). Thiol group exhibits high site-specificity for halide vacancies, also it tends to form robust Pb-S coordination bond. That effectively heals the ionic dangling bonds at the perovskite terminal facet. Amino groups provide bifunctional stabilization, involving both Lewis base-acid coordination with Pb²⁺ and hydrogen-bonding interactions with the halide. Both groups are designed to manage the perovskite interface. On the other hand, the oxidophilic phosphonic or carboxyl functional groups establish a robust, chemically tethered anchoring to the SnO₂ surface via coordination, ensuring superior mechanical stability and an intimate electronic junction. Figure 1 (b) show the calculated intermolecular electrostatic potential (ESP) of 3-mercaptopropionic acid (MPA), 6-mercaptopyridine-3-carboxylic acid (MPCA), and 2-aminoethylphosphonic acid (APA). In addition to functional groups, soft linear or rigid aromatic backbones were also discussed the effect of geometric effect on molecule orientation between interfaces.[32] The proposed molecule orientation between the interface is shown in Fig. 1 (c). Among them, APA has the lowest intermolecular ESP of -0.093 in this series. To realize the adsorption scenario of each molecule, their adsorption free energies onto SnO₂ surface were calculated and summarized in Fig. 1 (d). The negative adsorption energy of carboxyl group (-0.21 and -0.23 eV) and phosphate (-0.08 eV) reveal that only these two functional groups tend to adsorb onto the SnO₂ surface to maintain low free energy condition. As focusing on the perovskite interface, all molecules perform negative adsorption energy. The amine group on APA shows the lowest adsorption energy among them. Based on the above information, the MPA and APA molecules tend to coordinate linearly between SnO₂ and perovskite interface. That allows desired dipole moment to be created at SnO₂ and perovskite interface and paves a smooth electron transfer path from perovskite to SnO₂ layer.

Generally, the lattice spacing of SnO₂ (~ 3.3 Å) is significant smaller than that of the perovskite layer (~ 6.3 Å). This mismatch generates a stress-rich layer at the intimate surface, namely buried interface, of a perovskite layer. The resulting strain induces a disordered amorphous layer that deteriorates photovoltaic performance. It is believed that the alkyl-based linker provides rotational freedom between this interface. Such MILs serve as a geometrical tuner to reduce the lattice mismatch between a perovskite layer and an ETL and releasing the interface strain. To evaluate the tethered effect of MILs between an ETL and a perovskite layer, the perovskite layer deposited onto either pristine ETL or modified ETL and characterized by using both X-ray diffractometer and grazing-incidence wide-angle X-ray scattering (GI-WAXS). As illustrated in Fig. 2(a), the perovskite films generally exhibit a dominant (100) out-of-plane preferred orientation,[33] with the notable exception of the layer deposited on the MPCA-modified ETL. This divergence is possibly from the high planarity and inherent π - π stacking of aromatic structure in MPCA. The aromatic structure functions as a soft template during the initial stages of film growth, reorienting the perovskite nuclei and disrupting the standard texture observed with the other modifiers.[34] To gain insight the stress effect on deposited perovskite layers, XRD patterns reduced from GI-WAXS were summarized into Fig. 2 (b) to (e). The different incident angles of X-ray, φ from 2.5° (surface) to 5.0° (buried interface), reveal the depth-dependent information about perovskite layers. The reduction diffraction pattern from GI-WAXS show a profound peak shift corresponding to perovskite layers deposited onto different ETLs. GI-WAXS based 2θ - $\sin^2 \psi$ method was practiced to realize the scenario of intimate layers and to evaluate the corresponding stress and strain condition.[35, 36] As shown in **Figure S3**, the positive slope represents the compressive stress in perovskite layers. It is well-known that the stress-induced strain in a lattice causes a profound band structure evolution.[37] The

introduction of MIL molecules successfully alleviated compressive stress, which decreased from -18.47 GPa (on pristine ETL) to -10.59 GPa (on MPA-modified ETL), -17.01 GPa (on MPCA-modified ETL), and -14.81 GPa (on APA-modified ETL). This mitigation allows the perovskite layer to maintain its intrinsic band structure with minimal lattice interference. Azimuthal stress analysis further reveals that in-plane stress—representing horizontal effects—is most severe on pristine ETLs at -50.34 GPa. APA molecules successfully mitigated this to -37.46 GPa, a reduction of over 20%. In contrast, out-of-plane (vertical) stress remained relatively constant at -16.00 ± 2.00 GPa across all perovskite films as shown in **Figure S4 (a) to (d)**. This suggests that stress release occurs naturally during crystallization as film thickness increases.

From macroscopic points of view, surface roughness and morphology are critical for the sequential stacking of device layers. As shown in **Figure S5**, the average grain size of the perovskite remains consistent at around 330 nm deposited on both pristine and modified ETLs. It suggests that these MIL molecules have a negligible influence on the crystallization stage. However, topographies from atomic force microscopy (**Figure S6**) reveal a refinement in surface roughness (R_a). The R_a values decrease from 21.9 nm (pristine) to 18.6 nm (MPA) and 16.2 nm (APA), an improvement attributed to the successful release of interfacial stress. Such crystallization, interfacial dipole, and morphological improvements tantalize us to dig into the relationship between them and photovoltaic performance. Preliminary performance evaluations of 10-mM MILs (**Table S1**) identify APA-modified ETLs as the most promising candidates, exhibiting significant enhancements in V_{OC} . Consequently, pristine SnO_2 and APA-modified ETLs were selected as the control and target for microscopic point of view investigation.

To elucidate the electronic evolution at the heterojunction, the static XPS and *in-situ* XPS were conducted to observe the electrons evolution and transition between the ETLs and perovskite layers. The upward binding energy shift (0.8 eV) in Sn $3d$ and O $1s$ orbital of target ETL manifests a significant reduction in electron density as shown in Fig. 3 **(a)** and **(b)**. This electronic depletion confirms the successful anchoring of the electron-withdrawing phosphonate groups onto the SnO_2 surface. Conversely, the reverse scenario can be observed from the perovskite film deposited onto an APA-modified ETL. the perovskite film on the target ETL shows a downward shift in Pb $4f$ and I $3d$ binding energies (Figs. 3 **(c)** and **(d)**), signaling an increase in electron density mediated by the electron-donating amino groups. The *in-situ* observation under light illumination (Fig. 3 **(e)** and **(f)**) provides direct evidence of a highly efficient charge transfer pathway for perovskite film deposited onto target ETL. Despite the photo-generation of carriers, the perovskite film on the target ETL maintains a lower electron density (upward binding energy shift) compared to the dark state (Fig. 3 **(c)**). That confirms the interfacial dipole and the molecular bridge accelerate electron injection from the perovskite layer to the SnO_2 . Taking these advantages, the devices photovoltaic performance based on APA-modified ETL were expected to have promotion compared to the device made of pristine ETL. The preliminary tests of APA-modified ETLs were summarized in **Table S2** and the corresponding $J-V$ curves of champion devices were shown in **Figure S7**. While the bifunctional APA modified interface effectively mitigates interfacial strain and suppresses the potential barrier, its intrinsic dielectric character imposes a kinetic bottleneck at concentrations exceeding 3.0 mM. Beyond this threshold, the increased molecular layer thickness impedes charge carrier tunneling. This physical impedance consequently triggers a slight attenuation in overall device efficiency, highlighting the necessity of precise molecular density control to balance dipole-assisted extraction with tunneling resistance.

To further observe and validate the carrier dynamics under operating conditions, steady-state photoluminescence (PL) and time-resolved photoluminescence (TRPL) measurements were performed on perovskite films deposited onto either control or target ETL. As shown in Fig. 4 **(a)**, the perovskite onto target ETL exhibit significant PL quenching compared to the control. This suppression of radiative recombination is attributed to the presence of the interfacial dipole and the strong electron-withdrawing character of the phosphonate functional group, which effectively lowers the charge injection barrier. Furthermore, TRPL spectroscopy, Fig. 4 **(b)**, reveals a remarkable reduction in averaged carrier lifetime (τ_{eff}) for the target device with specific values detailed in **Table S3**. Through bi-exponential decay model, the carrier dynamic in a perovskite layer can be categorized into a fast decay component, namely charge transfer, and a slow decay component, radiative recombination.[38] While the ratio between these mechanisms remains relatively constant, the charge transfer time exhibits a profound reduction from 4.28 ns to 2.78 ns, representing a 35% improvement. This faster decay kinetics provide direct evidence of accelerated electron extraction and a streamlined charge transfer pathway facilitated by the linear orientation of the APA molecules. By simultaneously passivating surface trap states and inducing a dipole-assisted injection, the APA molecule effectively minimizes energy loss at the heterojunction. The V_{OC} of the device is fundamentally governed by the built-in potential difference based on energy band alignment. To evaluate the energy alignment scenario in an entire device, the valence band X-ray photoelectron spectroscopy (VB-XPS) were conducted to characterize the ETLs

and perovskite layer in **Figure S8** and summarized in Fig. 4 (c). The upshift conduction band minimum of target ETL results in an enlargement of the QFLS between the ETL/perovskite and perovskite/HTL interfaces. This increased QFLS is a primary driver for the observed V_{OC} enhancement. Statistical PCE analysis of fifteen individual devices (Fig. 4 (d)) reveals that the all devices show high reproducibility. The averaged PCE of target devices enhance to 21.81% as compared to 19.68% of the control devices. The corresponding J - V curves and external quantum efficiency (EQE) spectra of champion devices were shown in Figs. 4 (e) and 4 (f). With effective carrier dynamics, the PCE of champion devices can be promoted from 20.32% to 22.92%. The averaged EQE of device with target ETL in visible light region achieves 82.5% as compared to 80.0% of device with control ETL. The current density difference between control and target champion device is consistence with EQE difference of 2.50% enhancement. Based on the above characteristic information, the PCE enhancement is attributed to the potential difference enlargement and fast charge extraction between layers.

The enhancement in charge transfer efficiency is further corroborated by Contact Potential Difference (CPD) characterization under various light sources (Fig. 5(a)-(d)). Perovskite layer deposited onto target ETL performs large CPD change under different light sources on/off switching test. That refers that the electron separation is much effective in such perovskite layer. The minor carrier quenching in perovskite layers deposited on target ETLs perform much larger potential difference than that of control ETLs. As irradiation light energy closes to energy band gap of a perovskite layer, namely red light, the defect-assisted recombination process would be magnified. The decrease of CPD is attributed to the defect-assisted recombination. The perovskite onto target ETL show more different CPD under UV, blue, green, and red light irradiation. It again reflects that the MIL at interface can mitigate interfacial defects impact during carrier transport.

Furthermore, APA modification not only enhances photovoltaic performance but also significantly improves device stability. As shown in Fig. 6(a), the normalized PCE of unencapsulated the target device under ambient conditions retains 89% of its initial value for over 1,500 h, demonstrating excellent long-term stability. Under continuous light soaking conditions, the initial PCE of the target device can retain 80% lifetime of the modified device for 70 hours as compared to 25 hrs of the control device, as shown in Fig. 6(b). Additionally, the wettability of the perovskite surface is improved following APA modification, as evidenced by an increase in water contact angle from 63° to 92°, as confirmed in **Figure S9**. That makes moisture hard to penetrate into the perovskite layer deposited onto target ETL and prolong the device lifetime as well as reliability.

Conclusion

By integrating 2-aminoethylphosphonic acid (APA) as a heterobifunctional linker, we demonstrate a novel concept for harmonizing the SnO_2 /perovskite interface through simultaneous mechanical stress relief and electronic potential modulation. The bifunctional architecture of APA allows the electron-withdrawing phosphonate groups to anchor robustly onto the SnO_2 surface—evidenced by an upward binding energy shift of 0.8 eV—while the electron-donating amino groups passivate lead cations and halide vacancies in the perovskite lattice. This directional assembly establishes a well-defined interfacial dipole ($\text{ESP} = -0.093$) that lowers the charge injection barrier and expands the quasi-Fermi level splitting, resulting in a 35% acceleration in charge transfer kinetics and a champion PCE of 22.92%. Furthermore, the flexible alkyl backbone of APA acts as a geometrical tuner that alleviates compressive strain by over 20%, reducing in-plane stress from -50.34 GPa to -37.46 GPa to preserve the intrinsic perovskite band structure. The flattened and compact surface morphology of the perovskite film on target ETL exhibit high moisture persistence with 92° of contact angle. This synergistic effect not only enhances performance but also drastically improves device reliability. The devices made from target ETL can retain their 80% of initial PCE from 25 to 70 hours, thereby providing a robust blueprint for high-efficiency, durable perovskite photovoltaics.

Declarations

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Author Contribution

Hong-Yi Lin: Writing – original draft, Investigation, Formal analysis, Data curation. Kai-Chi Hsiao: Writing – original draft, Conceptualization, Investigation, Formal analysis, Data curation. Ting-Han Lin: Investigation, Data curation. Jia-Mao Chang: Investigation, Formal analysis. Wei-Chen Chu: Formal analysis. Wei-Hao Chiu: Investigation. Kun-Mu Lee: Resources. Ming-Chung Wu: Supervision, Resources, Project administration, Funding acquisition.

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Data Availability

All data supporting the findings of this study are available within the paper and its Supplementary Information.

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Figures

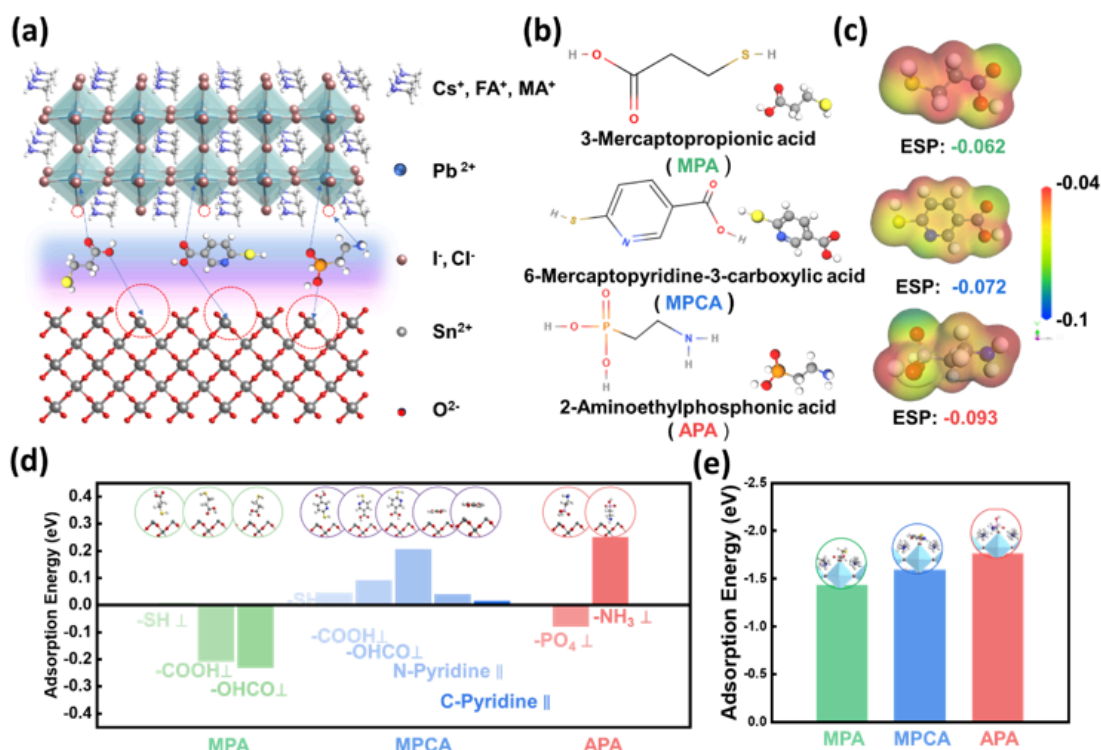


Figure 1

Interaction of MIL molecules between SnO_2 and perovskite: (a) molecule structures, (b) electrostatic potential of MIL molecules, and (c) illustration of MIL molecules at SnO_2 and perovskite interface. Calculated adsorption free energy of MIL molecules at (d) SnO_2 surface and (e) at perovskite surface with different ending functional groups.

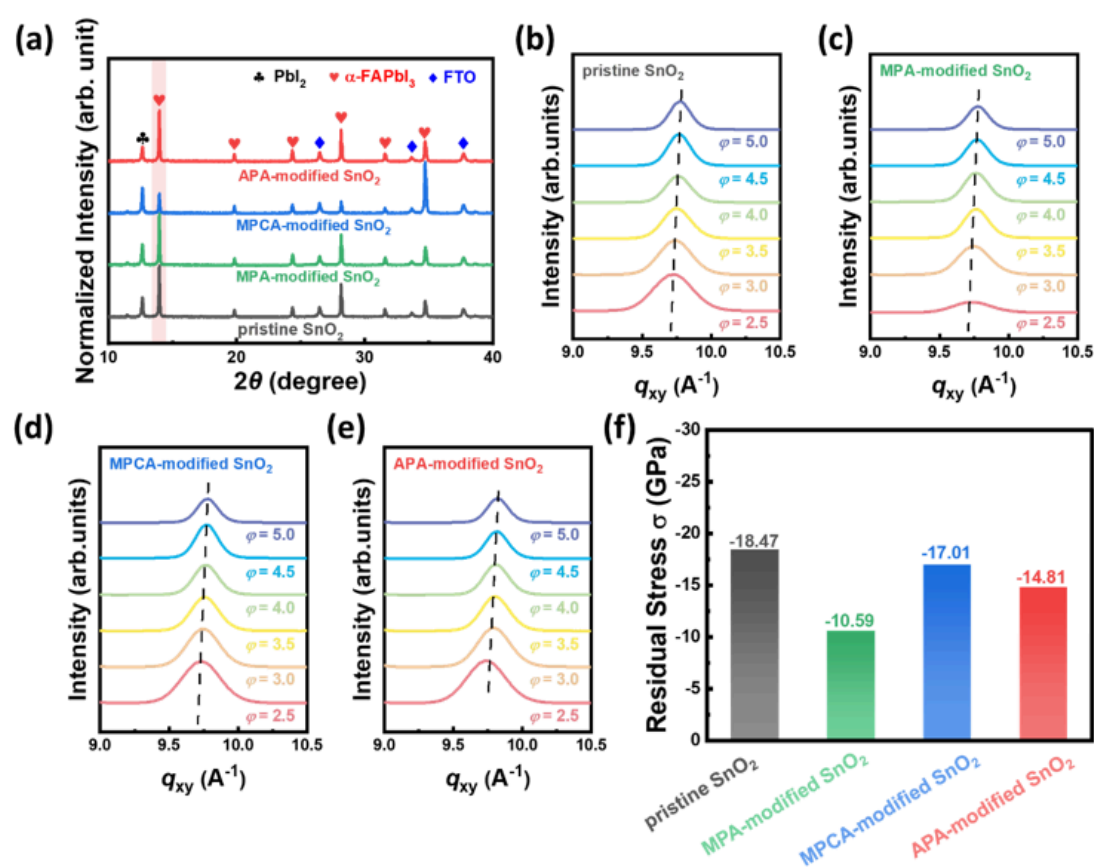


Figure 2

Characterization of perovskite film deposited onto different substrates: (a) XRD patterns of perovskite films on different MIL molecules. Depth-dependent lattice deformation of perovskite on (b) pristin SnO_2 , (c) MPA-modified SnO_2 , (d) MPCA-modified SnO_2 , (e) APA-modified SnO_2 , and (f) the corresponding residual compressive stress in perovskite films.

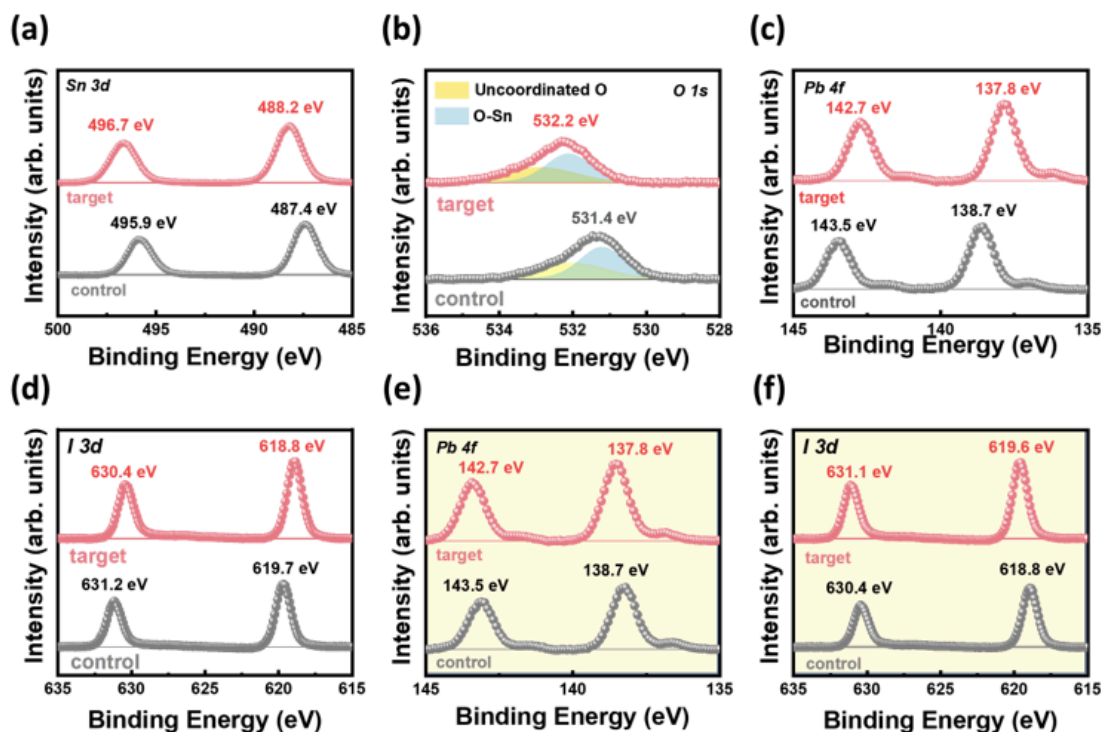


Figure 3

Oxidation state analysis: (a) Sn 3d orbital and (b) O 1s orbital of control or target ETL. *In-situ* XPS of (c) Pb 4f orbital, (d) I 3d orbital of perovskite deposited onto control and target ETL in dark, (e) Pb 4f orbital, and (f) I 3d orbital of perovskite deposited onto control and target ETL under illumination.

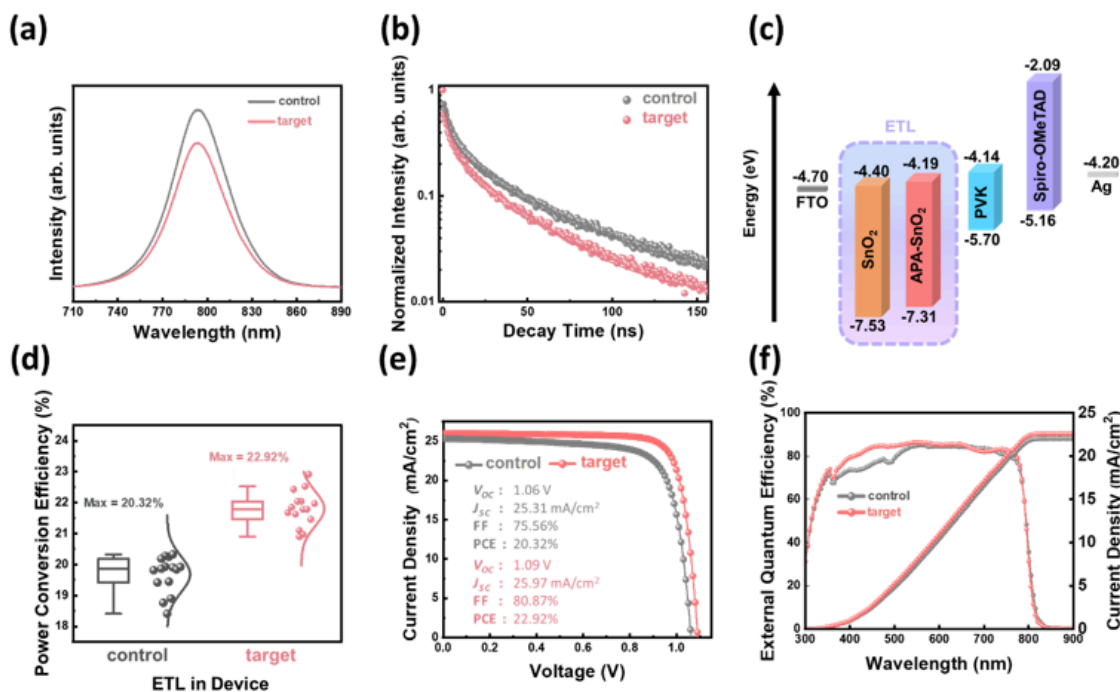


Figure 4

Photovoltaic performance of perovskite on control and target ETL: (a) photoluminescence spectra, (b) time-resolved photoluminescence spectra, (c) band diagram, (d) statistic PCE, (e) J-V curves, and (f) EQE spectra of champion devices.

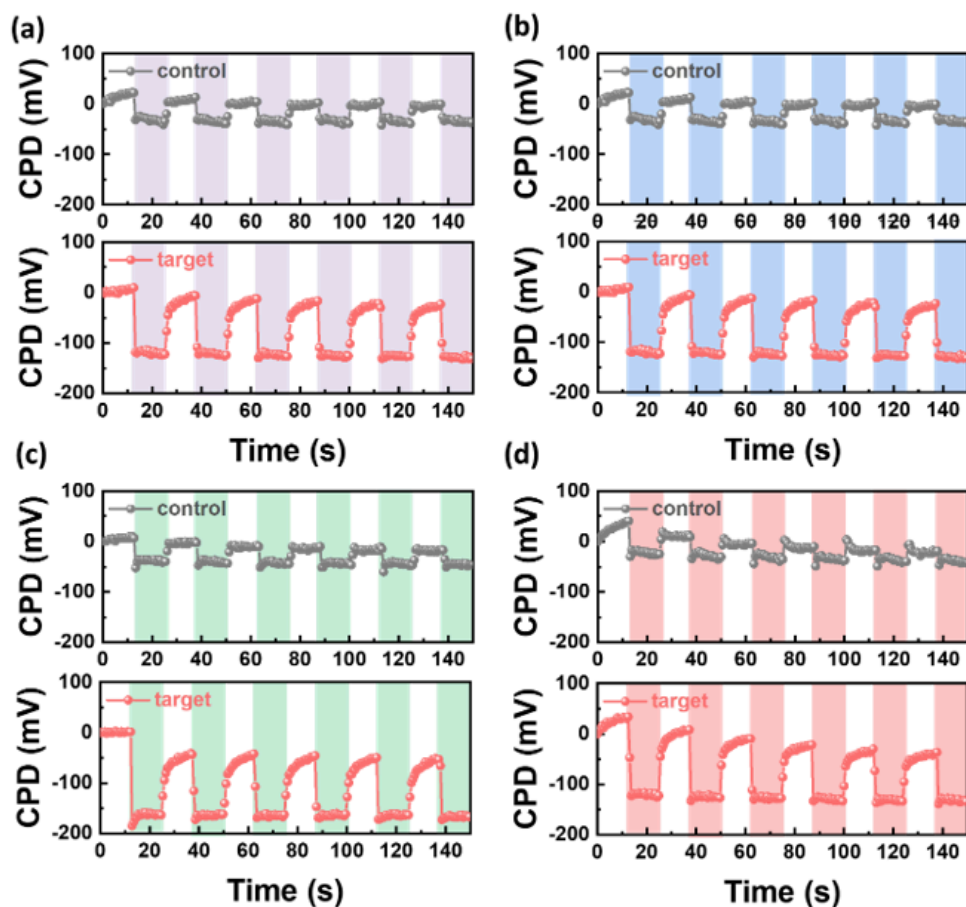


Figure 5

Contact potential difference of control or target device under: (a) UV-A, (b) blue light, (c) green light, and (d) red light irradiation.

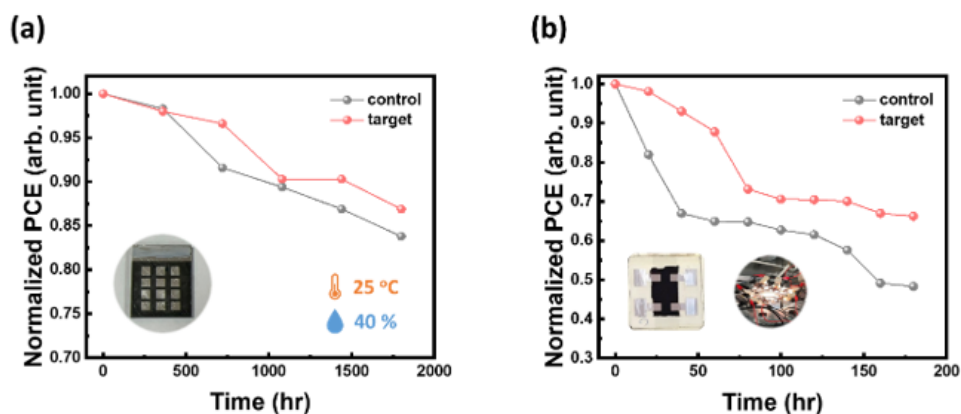


Figure 6

Long-term stability test of devices comprised of control and target ETL: (a) devices stored in air with 40 RH% at room temperature and (b) continuous light-soaking of devices without encapsulation.

Supplementary Files

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