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Self-Assembled Monolayers as Hole Transport Layer in Inverted Perovskite Solar Cells

Victorson Rabha, Basanta Kr Saharia

Department of Physics, Tangla College, Tangla

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ABSTRACT

SAMs have become one of the most essential materials in the modern inverted PSCs, setting the stage for molecular engineering at interfaces through simple scalable processing. In this review, we summarize the advancements that exploit SAMs as hole transport layers in the inverted (p-i-n) PSCs, addressing the outstanding challenges of molecular packing, adhesion to metal oxides, and wetting from polar precursors of the perovskite layer. We will highlight seven recent works from Chen et al., Li et al., Du et al., Ahmmed et al., Wang et al., Yuan et al., and Huang et al. for passivating surface defects with ligands, changing the core of the SAM to improve its dipole and binding energy, coupling SAMs to polymer or carbonaceous layers to exploit their advantages and compensate for their weaknesses, and replacing incompatible carbazole-based anchoring groups with stronger ones. All these strategies have delivered PCEs beyond 26% and meaningful operational stability improvements under accelerated aging. In this Review we describe what worked, what didn't and argue that, rarely, a single fix solves all the problems. Rather the most effective interventions often use a combination of approaches. We conclude the review with some thinking about where the field is currently challenged, and where future gains are likely.

Index Terms: inverted p-i-n PSC, optoelectronics, self-assembled monolayer perovskite

INTRODUCTION

Hole transport layers (HTLs) made from self-assembled monolayers (SAMs) have been popular in recent years for high-performance inverted perovskite solar cells (PSCs). The recorded power conversion efficiencies (PCE) of SAM-based inverted PSCs have exceeded 26% [1], [2], [3]. SAM molecules naturally arrange into a highly ordered layer that is only a few molecules thick. SAMs prevent charge accumulation at the HTL/perovskite interface, thereby significantly improving hole extraction from the perovskite by reducing interfacial electrical resistance [4]. One of the main advantages of SAMs is their structural tunability. SAM molecules contain a terminal group, a spacer, and an anchoring group. One can tune the work function of a transparent conductive oxide or electrode by modifying the molecules in the SAM. This induces a constant dipole moment at the interface and increases the open-circuit voltage (V_{oc}) [5]. The strong anchoring groups of SAMs form robust covalent bonds with the metal oxide substrate, which can easily withstand chemical processing techniques [4]. The anchoring groups of SAMs exhibit a strong binding affinity that can nullify the structural defects underlying the surface of the metal oxide and the perovskite layer. This reduces the defect density and the non-radiative recombination of charge carriers [5]. Due to its low cost and high scalability, SAMs became a popular choice among researchers [4].

Although SAMs have numerous advantages in inverted PSCs it also possesses certain limitations and engineering challenges. Solution-processed SAMs are susceptible to molecular aggregation, which causes the molecules to form clusters instead of forming a uniform monolayer. This can expose the underlying metal oxide layer to the perovskite layer and induce non-radiative recombination and interfacial energy loss [6]. SAMs containing highly acidic anchoring groups can corrode the metal oxide layer over time and degrade the potential

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stability of the device [7]. Some of the widely used SAMs possess hydrophobic terminal groups. Since the perovskite precursor solutions are highly polar, it strives to spread uniformly across the hydrophobic surface [8].

Review Study

Chen et al. (2024) have implemented ligands to passivate the surface defects of the perovskite [9]. They used BZS (benzenesulfonate) based ligands with functional groups containing methyl (CH_3) and chloride (Cl). Out of all ligands, they found that BZS with a Cl functional group (4Cl-BZS) was the most energetically favourable in parallel configuration to the perovskite surface. Due to the Cl functional group, the ligand binds with the Pb^{2+} defect sites at the surface of the perovskite and passivates it. $4CH_3$-BZS was found to be more energetically favourable in perpendicular orientation [9]. The researchers have fabricated BZS-based ligands in the perovskite precursor thin film using the spin coating method. They have used $Cs_{0.05}FA_{0.95}MA_{0.1}PbI_3$ as the perovskite material, where FA is formamidinium, and MA is methylamine.

The XPS (X-ray Photoelectron Spectroscopy) data showed that the binding energy of BZS-treated perovskites is lower than that of untreated films. This study suggested that Pb2+ ions underlying at the perovskite surface remain undercoordinated in the untreated films. The shift in the XPS peak in target films depicts the increment in electron density around the Pb atoms, which indicates the coordination of the ligand with the Pb^{2+} defect sites. The interaction of Cl and Pb atoms is further confirmed via NMR [9].

The PLYQ (Photoluminescence Quantum Yield) data revealed that the electron loss is minimal at the perovskite/C_{60} (ETL) interface of 4Cl-BZS treated fully stacked device [9]. The fully stacked device is fabricated as FTO/SAM/perovskite/C_{60}. FTO is fluorinated tin oxide and the SAM used is a mixture of and Me-4PACz. 2PACz is 2PACz (2-(9H-carbazol-9-yl)ethyl)phosphonic acid and Me-4PACz is [4-(3,6-Dimethyl-9H-carbazol-9-yl)butyl]phosphonic acid.

The TRPL (Time-Resolved Photoluminescence) data showed the average electron lifetime of 4Cl-BZS treated perovskite films is 3 μs, whereas the untreated control films had only 0.6 μs [9].

4Cl-BZS treated perovskite exhibited faster photocurrent decay as compared to $4CH_3$-BZS treated films [9]. This proves that charge extraction is faster is the former one.

Chen et al. (2024) have constructed the BZS-based PSCs as FTO/SAM/perovskite+ligand/C_{60}/SnO_x/Ag. FTO and Ag are the electrodes, SAM is the HTL, C60 is the ETL, and SnO_x is an interfacial layer that enhances electron extraction from the ETL to the Ag cathode [9].

The power conversion efficiency (PCE) of the champion device is 26.3%. Chen et al. (2024) also added a BMP (Biomolecular Surface Passivation) layer in the 4Cl-BZS treated perovskite [9]. This BMP layer contains two molecules, 3MPTAI and $PDAI_2$. 3MPTAI stands for 3-Methylthio-1-propylammonium iodide and $PDAI_2$ is Propane-1,3-diammonium iodide. The combined treatment of the BMP layer and 4Cl-BZS ligand helped the PCE to increase from 26.3% to 26.9%. This device yielded an open-circuit voltage, V_{oc} of 1.18 V, FF (Fill Factor) of 86.2% and short-circuit current density, J_{sc} of 26.4 mA/cm^2 [9].

Under protocol ISOS-L-3, the target device managed to retain 95% of its initial efficiency at maximum power point (MPP) after ageing 1200 hours under 1-sun illumination at 65°C (at 50% RH) [9].

Li et al. (2023) have utilized MeO-4PADBC and NiO_x as the HTL in designing high-performance p-i-n PSCs based on SAM. MeO-4PADBC stands for (4-(3,11-dimethoxy-7H-dibenzo[c,g]carbazol-7-yl)butyl)phosphonic acid. This molecule contains a dibenzo[c,g]carbazole (DBC) core, which is non-coplanar and asymmetrical [10]. It prevented the OMe groups from cancelling each other; as a result, it exhibited a strong dipole moment. A strong dipole moment assists in effective hole extraction from the perovskite by inducing Ohmic contact between the metal oxide and perovskite layer. A combination of NiO_x and MeO-4PADBC helps the device achieve low defect density [10]. The thermal degradation energy of this HTL is almost 3 times that of standalone MeO-4PADBC.

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The binding energy between the SAM and the perovskite was found to be -7.19 eV [10]. The strong binding is possible due to the interaction between O atoms from the OMe group and Pb from the perovskite.

The fully stacked device was constructed as follows: ITO (Indium Tin Oxide)/HTL/perovskite/C_{60}/BCP/Ag. BCP(Bathocuproine) is an interfacial layer between C_{60}(ETL) and Ag(cathode) [10]. Li et al. (2023) have used $Cs_{0.05}FA_{0.85}MA_{0.1}PbI_3$. BCP layer blocks holes from passing from the ETL to the cathode. They have also fabricated a 2D passivation layer between the ETL and perovskite. The 2D passivation layer contains CF3-PEAI (4-Trifluoromethylphenylethylammonium iodide) and MAI (Methylammonium iodide) in a 3:4 ratio [11]. This layer passivates the undercoordinated Pb ions and halide vacancies underlying on the surface of the perovskite.

NiO_x/MeO-4PADBC based PSCs having a band gap of 1.53 eV yielded an average PCE of 25.6% for a mask area of 0.0414 cm^2. It showed V_{oc} = 1.19V, J_{sc} = 25.4 mA cm^{-2} and FF = 82.1%. For 1.68 and 1.80 eV band gap target devices, the PCE was found to be 22.7 and 20.1%, respectively [10]. It can clearly be seen that the PSC with 1.53 eV band gap is the most efficient one.

Li et al. (2023) have conducted thermal accelerated ageing tests to study the stability of the PSCs under real-world conditions. The devices were treated under 1-sun illumination with a fixed load resistance near the MPP (Maximum Power Point) with temperature ranging from 25°C to 100°C, following the ISOS-L-2I protocol [12]. The NiO_x/MeO-4PADBC based devices maintained the PCE at 90% of its initial efficiency after ageing for 1200 hours at 65°C and 74% at 85°C [10]. From the results of the lifetime acceleration factor (AF), they estimate that the target device could retain 80% of its initial PCE at room temperature after 7567 hours of ageing[10].

Du et al. (2026) have implemented printable carbon paste as the cathode via the polarity inversion method [13]. Typically, carbon exhibits hole selectivity. It is generally used as an anode in n-i-p PSCs [14]. The research has been conducted to invert the polarity of carbon from anode to cathode.

They developed an SnO_x interlayer between the ETL and the carbon cathode. This interlayer is deposited via the ALD (Atomic Layer Deposition) method; hence, the interlayer is called ALD-SnO_x[13]. The ALD-SnO_x layer is highly uniform, dense and amorphous. Due to its robustness, this interlayer prevents the carbon paste from penetrating it. The optimal thickness of this ALD-SnO_x layer is about 5 - 25 nm [13].

Annealing the ALD-SnO_x layer under UV radiation and in the presence of N_2 induced oxygen deficiencies in the film. This process is called self-doping, by which the ALD-SnO_x layer is tuned as an n-type layer [15]. Self-doping the ALD-SnO_x layer is essential since the amalgamation of undoped SnOx and Carbon induces a Schottky contact at their interface. By self-doping, the thickness of the depletion region decreases; as a result, the electrons can tunnel through the Schottky barrier [16].

Du et al. (2026) have constructed the device as: ITO/HTL/perovskite/ETL/SnOx/Carbon. ITO is Indium-doped Tin Oxide. HTL layer is composed of SAM (NIOx/Me-4PACz), and C_{60} have been used as the ETL. They utilized $Cs_{0.055}MA_{0.047}FA_{0.898}PbI_{0.953}Br_{0.047}$ as the perovskite material. It is found that the ALD-SnO_x layer should have a thickness in the range 5 – 25 nm for effective performance of the device [13].

The target device achieved a PCE of 21.8% [13]. Carbon is a chemically inert element; hence, it eliminates electrode-induced degradation [17]. Applying accelerated thermal and light ageing under the ISOS-L-2I standard, the SnO_x/Carbon based PSCs revealed its activation energy is about 0.61 eV. Du et al. (2026) predicted the T_{80} lifetime at 30°C and found the target devices could maintain 80% of its initial PCE for 8240 hours [13]. Carbon is a cheap and abundant material which makes SnO_x economically viable.

Ahmmed et al. (2026) have synthesized triphenylamine based conjugated SAMs and functionalized different numbers of methoxy groups in different positions of triphenylamine. They investigated its effects on the perovskite layer and the PSC [18]. Out of all the SAMs, they found C22-based PSCs to be the most efficient with a PCE of 21.58% [18]. They configured the device as: FTO/SAM/perovskite/C_{60}/BCP/Ag. The perovskite material they used is $Cs_{0.05}FA_{0.80}MA_{0.15}PbI_{2.75}Br_{0.25}$.

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The C22 molecule has methoxy groups (-OCH_3) at both ortho and para positions of the triphenylamine core molecule. This molecule utilizes the conjugated linkers to connect the core to the functional group. It promotes electron delocalization and provides a less-resistive pathway for charge carriers [19], [20], [21], [22]. The symmetricity of the C22 molecule suppresses the molecule from aggregating in the precursor solution. This promotes the growth of a dense and conformal layer of SAM between the FTO and perovskite layers [18].

Depositing C22 in the FTO shifts the work function of the FTO to -4.76 eV. This phenomenon induces upward band bending, which accelerates the hole transfer rate. The methoxy groups located at the para position of the C22 molecules directly point toward the perovskite [18]. These methoxy groups passivate the undercoordinated Pb^{2+} ions located on the surface of the perovskite. As a result, the defect density of the C22 molecule is reduced to 3.10×10^{15} cm^{-3}. The C22-based PSC yielded V_{oc} = 1.07V, J_{sc} = 24.12 mA cm^{-2}, FF = 83.63%, and PCE = 21.58% [18].

Wang et al. (2026) have developed PTAA and SAM at the FTO substrate as HTL [23]. PTAA is referred to Poly[bis(4-phenyl)(2,4,6-trimethylphenyl)amine]. The roughness of the FTO surface is about 150 nm. Covering this rough surface with solution-processed PTAA becomes extremely thick. The highly thick PTAA layer reduces shunt resistance but increases series resistance [23]. On the other hand, SAMs have high hole selectivity, faster hole transfer rate and low interfacial trap density [24], [25], [26]. SAM can anchor to metal oxide substrates; however, the anchoring is not strong enough to form a uniform and dense single monolayer [27], [28].

PTAA and SAM are highly compatible to each other. SAM allows the PTAA layer to be thinner and provides low series resistance. Wang et al. (2026) deployed MeO-2PACz as the SAM. Also, the PTAA: SAM composite layer exhibits lower surface energy inhomogeneity [23]. PTAA is deposited at the valleys of the FTO substrate, while the SAM is anchored at the peaks of the substrate, where the reach of PTAA is minimal [23].

The p-i-n PSC is structured as: FTO/HTL/F127/perovskite/PEAI/PCBM/C_{60}/BCP/Ag [23]. F127 is Poloxamer 407, PEAI is phenethylammonium iodide, and PCBM is [6,6]-phenyl-C_{61}-butyric acid methyl ester. The device yielded average PCE = 22.7%, V_{oc} = 1.09V, FF = 82.1%, and J_{sc} = 24.8%. The improvement in the PSC performance is due to controlled series resistance and largely increased shunt resistance. Applying non-ionic copolymer surfactant F127 as the interfacial layer increases the PSC to 25.4% [23].

The unencapsulated devices were tested under AM 1.5 g-1 sun at 30°C. The test results revealed that the PTAA: SAM-based devices managed to retain 100% of its initial PCE after ageing for 1000 hours [23].

Yuan et al. (2025) have utilized TPA2P as the HTL material for the p-i-n PSC. They incorporated a conjugated bisphosphonic acid anchoring group in the SAM [3]. TPA2P is the SAM molecule, which stands for (2-(4-(diphenylamino)phenyl)-1- phosphonovinyl)phosphonic acid.

The bisphonic acid configuration strengthens the binding on the indium tin oxide (ITO) substrate, which improves the uniformity and interfacial stability of the SAM [3]. The ethyl group located in the TPA2P molecule enhances the intermolecular charge transfer (ICT) from the triphenylamine core (electron-donating) to the bisphosphonic acid group (electron-accepting). The charge redistribution induced by the ICT causes the HOMO (Highest Occupied Molecular Orbital) level of the SAM to shift to -5.47 eV. Due to the optimization in energy alignment, the interfacial energy loss is reduced, improving the efficiency of hole extraction [3].

Yuan et al. (2025) fabricated the following PSC: ITO/TPA2P/$Cs_{0.05}FA_{0.95}$/C_{60}/BCP/Ag. The SPO of the device showed that the PCE is in the range 25.61 - 26.08%. The optimized TPA2P-based devices revealed V_{oc} = 1.18 V, FF = 85.03%, and J_{sc} = 25.99 mA cm^{-2} [3]. The TPA2P-based devices were tested at 40 ± 5°C, AM 1.5G illumination with N_2 atmosphere, and the ageing test results revealed that the devices retained 96.7% of its initial PCE after ageing for 1000 hours [3].

Carbazole-based phosphonic acids such as 2PACz and Me-4PACz are among the most widely used SAMs [10], [11], [26]. But these carbazole-based SAMs lack the adhesive binding strength to attach to the transparent conducting oxides and perovskite layer [29], [30], [31]. To overcome the weak binding issue of carbazole-based SAMs, Huang et al. (2025) have developed PAFTB [32]. PAFTB stands for 4-(7-(4 (bis(4-

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methoxyphenyl)amino)-2,5-difluorophenyl)benzo[c][1,2,5]thiadiazol 4-yl)benzoic acid. PAFTB exhibits both donor and acceptor properties.

PAFTB has a higher dipole moment (4.93D) as compared to 2PACz (1.97D) [32]. This improves hole extraction at the HTL/perovskite interface. PAFTB accommodates numerous functional groups such as carboxyl, methoxy, fluoro and thiadiazole. This provides both electron-rich and electron-deficient sites at the SAM lattice. The undercoordinated ions underlying at the surface of the perovskite can be passivated by these functional groups [32].

The results of Sandwich double-cantilever tests showed that the interfacial adhesion of PAFTB is 2.8 times that of 2PACz [32]. The binding energy of PAFTB in SnO_2 is found to be 4.01 eV, whereas for PACz it was found to be 3.41eV [32].

PAFTB-based devices showed a PCE of 25.6%, FF = 84.0%, V_{oc} = 1.16V, J_{sc} = 26.2mAcm^{-2}. The T80 lifetime is 900 hours at 85℃ [32].

CONCLUSION

In summary, we have reviewed seven recent advances on SAM-based HTLs for highly efficient inverted perovskite solar cells, which tackle the issues from different perspectives. Specifically, Chen et al. showed that ligand passivation (4Cl-BZS) and BMP bilayer give rise to an efficiency increase up to 26.9% and excellent thermal stability, while Li et al. demonstrated that the introduction of NiO_x and structural tuning of the MeO-4PADBC molecule improved the binding energy and defect suppression. Du et al. found that the SAM effect could be applied to a carbon cathode through an ALD-SnO_x interlayer on top of the HTL layer. Ahmmed et al. demonstrated that C22 triphenylamine SAMs of different molecular symmetries could suppress aggregation, yielding more uniform films. Amalgamagation of SAMs, such as in Wang et al. with SAM and PTAA, compensates for suboptimal substrate coverage and resistivity. To overcome the weak-adhesion of carbazole-based SAMs, Yuan et al. and Huang et al. used bisphosphonic and donor-acceptor anchoring groups, respectively, to improve interfacial stability without sacrificing performance.

None of ligand passivation, molecular engineering, hybrid architectures, or anchoring-group engineering alone has solved the problems of SAM-based HTLs; the most success has been achieved by combining strategies. At present, this material only works well in a controlled lab environment. Before making it commercially available, the device should be designed to be tough enough to survive real-world conditions and agree on the test based on its lifespan.

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