



Perspective

Charge-transport layer engineering in perovskite solar cells

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Photovoltaic (PV) technology that directly converts the solar energy into electrical energy, is regarded as one of the most promising utilization technologies of renewable and clean energy sources. Nowadays, developing low-cost and highly efficient PV technology is a hot research topic both for academia and industry. In this context, perovskite solar cells (PSCs) with metal halide perovskites $[\text{ABX}_3]$, $\text{A} = \text{CH}_3\text{NH}_3^+$ (MA^+), or $\text{CH}(\text{NH}_2)_2^+$ (FA^+), Cs^+ ; $\text{B} = \text{Pb}^{2+}$, Sn^{2+} ; $\text{X} = \text{Cl}^-$, Br^- , I^-) as light harvesting material, is in the spotlight due to its easy fabrication process and high power conversion efficiency (PCE) [1,2]. To date, the certified PCE has been already pushed up to 25.2% (<https://www.nrel.gov/pv/module-efficiency.html>), making PSC an auspicious candidate for a new generation of photovoltaics. In future days, how to eliminate the non-essential charge carrier recombination in the device, further push the PCE approaching the Shockley-Queisser theoretical efficiency limit (~33%) and enhance the device stability, will be formidable challenges and the focus in the next stage of research work.

From the development history of PSCs, it is found that the majority of the highly efficient PSCs, whether n-i-p structured or inverted p-i-n structured device, commonly possess typical layer-by-layer architectures. In all of these architectures, the charge carrier transfer processes at the interfaces, including the perovskite/electron transport layer (ETL) interface, perovskite/hole transport layer (HTL) interface, together with the interfacial materials, and the following charge carrier transport processes in the bulk of ETL and HTL, play crucial roles in achieving high PCE and good stability (see Fig. 1). Efficient charge carrier generation (process 1 in Fig. 1), extraction and transport (process 2 and 3 in Fig. 1) are necessary to obtain high PCEs. Suitable ETL and HTL can improve the charge carrier extraction and reduce recombination (process 4, 5, and 6 in Fig. 1). Herein, we briefly review the recent

advances and provide a perspective for charge transport layer engineering for highly efficient and stable PSCs. According to the function of charge transport layer, ETL and HTL engineering are divided and discussed separately.

Electron-transport Layer. The perovskite/ETL interface has been proved to be absolutely vital for the electron extraction, transfer and charge recombination dynamic processes, correspondingly, influencing the performance of PSCs. Some researchers proceed from the perspective of optimizing energy level alignment at perovskite/ETL interface to enhance the electron extraction and transport efficiency. To date, the common strategies to build the optimal energy alignment involve designing novel ETL with more suitable energy levels, tuning the energy levels via doping, and inserting an interlayer between ETL and perovskite layer.

As a typical electron transport material (ETM) for PSCs, TiO_2 plays an important role in electron extraction and transport process in device. A PCE of 24.66% has been achieved by using TiO_2 as ETL (detailed device structures and performance data are shown in the Supplementary materials) [3]. However, the high-temperature processing, intrinsic low electron mobility of TiO_2 , large energy barrier at the perovskite/ TiO_2 interface and deep traps induced by UV light inhibit efficient charge transfer, leading to serious energy losses and hysteresis. Replacing the TiO_2 with SnO_2 is reported to be an effective way to enhance the electron transfer from perovskite to ETL, and reduce electron accumulation at the interface due to the much deeper conduction band and higher electron mobility of SnO_2 . Another advantage of SnO_2 is that it can be prepared at low temperature (150 °C). Different preparation methods were reported, including diluting a commercialized colloidal dispersion, sol-derived nanocrystalline method, atomic layer deposition (ALD) method, and a two-step colloidal synthesis method proposed by Fang group [4,5]. For sol-derived nanocrystalline method, the SnO_2 crystalline is sensitive to annealing temperature. Low annealing temperature will result in relatively low electron mobility, while high annealing temperature may break down the film, leading to inferior performance. The SnO_2 film

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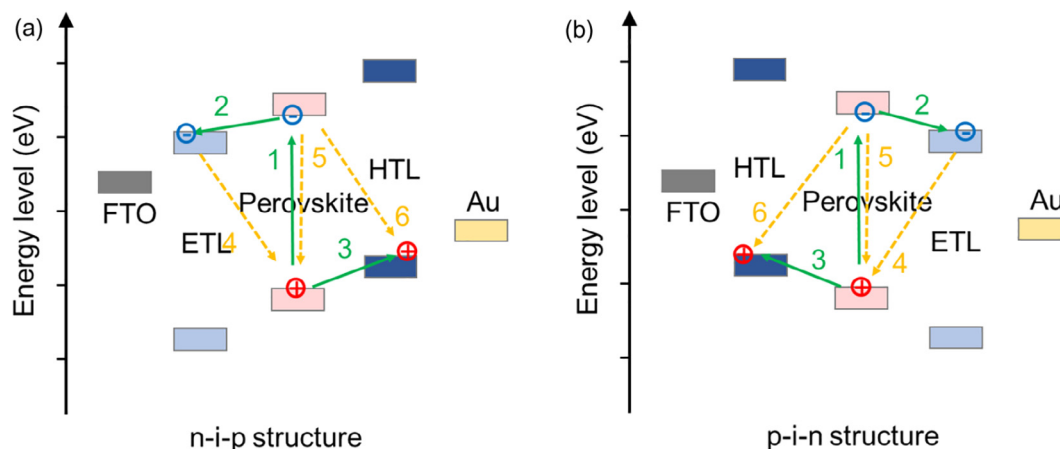


Fig. 1. (Color online) The typical charge carrier transfer process of (a) n-i-p and (b) p-i-n structured PSCs: 1. Photo-generation of electron-hole pairs in the perovskite absorber layer, 2. Electron extraction and transport process, 3. Hole extraction and transport process, 4, 5, 6 are undesired charge recombination processes taking place in working devices.

prepared with ALD method is compact but it is amorphous, therefore, the electron mobility is low. Colloidal synthesis method can overcome above dilemma, maintaining high crystallinity of SnO_2 nanoparticles and ensuring the compactness of SnO_2 film. To date, a PCE of 23.56% has been reported for perovskite solar cells with SnO_2 ETL [6]. Nonetheless, SnO_2 suffers from low wettability and lots of surface defects, and thus requires suitable surface modifications. Alternatively, BaSnO_3 was also manifested to be a promising ETL candidate for improving both the device PCE and stability [7]. Shin et al. [7] reported a low-temperature processed La-doped BaSnO_3 film as ETL to reduce the ultraviolet-induced damage to perovskite, and the fabricated PSCs can retain over 90% of the initial efficiency after 1000 h of full sun illumination. Similar research strategies were also implemented in the inverted (p-i-n) structured PSCs. Fullerene (C_{60}) and [6,6]-phenyl-C61-butyric acid methyl ester (PC_{61}BM) are the most prevalent ETMs for efficient inverted PSCs. Recently, a PCE of 23.0% was reported for C_{60} cells [8]. However, PC_{61}BM is not a perfect ETL because of the poor film-forming property, ordinary electron conductivity, and poor phase stability in ambient condition. Therefore, some new alternatives were developed, such as n-type conjugated polymer PDTzTI, which has been used in PSCs with a PCE of 20.86% [9]. Luo et al. [10] reported a PDI/fullerene hybrid supermolecule PDI-C60. Applied in PSCs, the supermolecule PDI-C60 achieves higher PCE of 18.6% and better device stability than the monomers (PC_{61}BM and PDI), due to the slightly shallower energy level, higher electron mobility and stronger hydrophobic properties of PDI-C60. Despite these successes, development of new organic ETMs with high electron mobility, suitable energy levels, and respectable photochemical stability remains a challenge.

Another effective way to promote the device performance is modifying well-developed metal oxide ETMs (TiO_2 , SnO_2 and ZnO) by using organic self-assembled monolayers (SAMs). Fullerene and its derivatives, such as PC_{61}BM , [6,6]-phenyl-C71-butyric acid methyl ester (PC_{71}BM), [6,6]-phenyl-C61-butyric acid-diethyl-3,3-(5-hydroxy-1,3-phenylene)-bis(2-cyanoacrylate) ester (PCBB-2CN-2C8), were successfully proposed as interfacial materials between ETL and perovskite because of their high electron mobility [11,12]. Along with fullerene derivatives, PDI derivatives and amino acid analogs were also proposed as interlayer materials. The SAMs not only can passivate the surface defects of metal oxides, but also can induce higher built-in internal electric field and dipoles pointing away from metal oxides, correspondingly, leading to better electron extraction and eventually achieving excellent photovoltaic performance. Very recently, two-dimensional graphi-

tic carbon nitride (g-CN) has also been explored as an efficient solution-processable interface modifier for the PC_{61}BM ETM to improve the device performance, suggesting a new interfacial engineering strategy [13].

Hole-transport Layer. As important as ETL, HTL is responsible for efficient hole extraction at the perovskite/HTL interface and hole transport in the bulk of HTL, and prevention of undesired charge recombination processes, contributing to outstanding photovoltaic performance. To get high PCE, hole transport materials (HTMs) with high hole mobility and sufficient conductivity, and cascade energy level alignment of the HTL with perovskite are both required. Moreover, transparent window in visible region is highly desirable for HTMs used in inverted and tandem structured PSCs. To ensure long-term stability, the HTL should be thermally and photochemically stable, as well as high hydrophobic to prevent from the degradation of perovskite by moisture and oxygen. Additionally, to realize low-cost device manufacturing, an ideal HTM candidate has to be easily developed by simple synthetic routes and easy purification procedures.

For n-i-p structured PSCs, Spiro-OMeTAD and PTAA are the two most employed HTMs. Although Spiro-OMeTAD and PTAA achieve great jobs in the charge separation and transport processes, for higher efficiency, better long-term stability and lower manufacturing cost, the development of new HTL and relevant interfacial engineering are still required to be further carried on. The first way is developing more efficient novel HTMs through integrating a variety of molecular engineering strategies, including optimization of core building block and alkyl chains, adjusting the electron-donating ability of peripheral groups and π -bridges. Due to the huge success of Spiro-OMeTAD during the past two decades, many pioneer research groups are devoted into developing Spiro-type HTMs through slight structural modifications. Many studies indicate that the methoxy triphenylamine (MOTPA) donor unit was unstable. To overcome this problem, Jeon et al. [14] reported a fluorene-terminated HTM DM with a slightly deeper lying HOMO energy level and a higher glass transition temperature, achieving a PCE as high as 23.2% and excellent thermal stability. Xu et al. [15] reported a series of spiro[fluorene-9,9'-xanthene] (SFX) core building block based HTMs. The SFX core unit can be easily synthesized by a one-pot approach with high yield, which is greatly helpful for pushing the price of HTMs down. The well designed SFX trimer X55 with excellent 3D configuration rendered an impressive PCE of 20.8% and greatly improved stability because of uniformity and homogeneity of the X55 film. Self-doping strategy and adjusting the planarity of molecular configuration are also manifested to

be effective paths to build impressive HTMs. Cheng et al. [16] reported a self-doped small molecular HTM by ionizing the end-capping pyridine group. The self-doping method can dramatically enhance the hole mobility and conductivity of HTM, and avoid the usage of LiTFSI and 4-*tert*-butylpyridine (TBP) in HTL, thereby simultaneously improving the PCE and stability. Based on a tetrathienylethene (TTE) core structure, Shen et al. [17] proposed a concept of hybridization between *planar* and *orthogonal* molecular conformation for balancing the charge mobility and thin-film quality of organic HTMs, which resulted in high performance dopant-free PSCs. Wang et al. [18] designed a methoxy-free donor alternative to MOTPA by fusing a fluorene to a carbazole, and developed HTM DM129 adopting the newly designed peripheral donor group. The introduction of dimethyl fluorene is found to improve planarity of molecule, film morphology uniformity, hole extraction efficiency and device PCE. Moreover, introduction of passivation functions into design of HTMs would also be a promising strategy to improve hole extraction and transport. Xiao et al. [19] recently developed a new class of polysquaraine-based HTMs with suitable energy levels, comprehensive passivation effects and high hole mobility close to $0.01 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. Benefiting from these merits, the fabricated dopant-free α -CsPbI₂Br-based PSCs delivered an impressive PCE of 15.5% and good stability. These reports suggest that tuning the molecular building-blocks, peripheral groups, end-capping groups and molecular configurations to improve performance and stability of PSCs is possible. Further studies addressing structure-function correlations are needed for developing novel HTMs to reach a real application level.

For p-i-n structured PSCs, poly(3,4-ethylenedioxythiophene): polystyrene sulfonate (PEDOT:PSS) has been the most widely used HTL at the beginning, and the cells gave PCEs as high as 20.1% [20,21]. However, the acidic and hygroscopic characteristics of PEDOT:PSS severely limit the long-term stability of devices. PTAA, a polymeric HTM widely used in normal structured PSCs, is also widely used as HTL in inverted PSCs, yielding a high efficiency of 23.0%, which is the record efficiency for inverted PSCs [8,9]. Besides, to overcome above shortcomings of PEDOT:PSS, lots types of inorganic p-type semiconductors, such as copper iodide (CuI), copper thiocyanate (CuSCN), copper oxides (Cu₂O and CuO) and nickel oxide (NiO) have also been developed as alternatives, due to the merits of easy fabrication, low production cost and suitable energy levels, and among which, NiO is the most prevalent one [22–26]. Jeng et al. [23] first applied a solution-processed NiO_x thin film in inverted PSCs. Later, Jung et al. [24] developed Cu-doped NiO for highly efficient PSCs. The combustion-processed Cu:NiO film showed enhanced electrical conductivity, resulting in improved PCE as high as 17.7%. Chen et al. [22] reported heavily p-doped NiMgLiO and achieved a PCE > 15% with an aperture area > 1 cm². Most recently, Huang et al. [27,28] proposed a series of ternary metal oxide (TMO) nanoparticles, such as In:CuCrO₂, ZnCo₂O₄ and NiCo₂O₄, as efficient HTLs in PSCs, demonstrating improved stability and high PCEs.

The other way is introducing inserting layer between HTL and perovskite, or constructing binary HTL to form a cascade energy level alignment, facilitating the hole transfer. Here, we have to note that the exact energy levels for a charge transport material could have a change when forming a close contact with the perovskite layer. Poly(3-hexylthiophene) (P3HT) is a promising potential HTM in PSCs; while the strong electronic coupling between the flat P3HT molecules and the perovskite, and the poor physically contact between P3HT and perovskite result in inferior performance. To improve the surface contact and to effectively reduce charge recombination at the perovskite/P3HT interface, Jung et al. [29] introduced a thin layer of wide-bandgap halide perovskite between the perovskite light harvesting layer and P3HT through an in-situ reaction of n-hexyl trimethyl ammonium bromide. The

optimized device showed a certified PCE of 22.7% and excellent stability. Ding group [30] developed an efficient binary HTL by blending traditional HTM PTAA with a polymer HTM PBD2T for CsPbI_{2.25}Br_{0.75} based PSCs. The cascade energy level alignment of CsPbI_{2.25}Br_{0.75}, PBD2T and PTAA greatly improved the hole extraction efficiency and reduced the unnecessary energy loss. Meanwhile, the S atom in PBD2T molecule is liable to form S-Cs and S-Pb bonds with bared metal atoms, passivating the surface defects of CsPbI_{2.25}Br_{0.75} and suppressing the interfacial charge recombination. CsPbI_{2.25}Br_{0.75} based PSCs with binary HTL delivered a PCE of 17.37%, which is the highest efficiency for Br-doped inorganic PSCs. Therefore, developing the cascade energy level alignment HTL system is another effective and promising strategy to fulfill the efficient hole extraction and transport.

In addition to the engineering of bulky properties of HTL, the interfaces between HTLs and the perovskite layers could also be directly tailored by designing the HTM molecules. Zhang et al. [31] constructed a dopant-free polymeric HTM DTB with 2,5-dialkoxy-1,4-phenylene units and bithiophene units. The intramolecular interaction between the oxygen atoms in the side chains of benzene rings and the sulfur atoms in the thiophene rings imparted a high coplanarity to the backbones of the polymers, leading to a high hole mobility for the HTL. On the other hand, the optimized length of the side-chains on benzene rings rendered the polymer adopt an edge-on arrangement atop the perovskite layer, thus benefiting a denser exposure of the thiophene rings to perovskite materials and the resulting hole extraction process. The two blank thiophene rings without any side-chains further led to a closer exposure of themselves and strengthened the ability of hole extraction from and defect passivation to the perovskite layer. As a result, a very high short-circuit current density of 25.75 mA cm^{-2} and an impressive efficiency of 19.68% were accomplished based on such a simple molecular structure. An optimization on the side-chains was conducted and part of them were replaced by polar ones later in order to increase the hole mobility of the polymer, and the corresponding PSC device yielded an improved PCE of 20.19% [32], further indicating the importance of engineering the bulky and interfacial properties of HTLs through the delicate molecular design.

In this perspective, we provided an outlook on the evolution and prospects of charge transport layer engineering for efficient and stable PSCs. We think, just with the crystal quality control and composition regulation of perovskite layer, it is insufficient to push the PCE approaching the theoretical efficiency, and achieve high stability. Great efforts should also be devoted to charge transport layer engineering to improve the device reproducibility and stability, and reduce device cost. Although a various of charge carrier transport materials and modification approaches have been proposed, the studies of charge transport layer engineering still need to be propelled. With the reported charge-transport materials, PSCs have achieved great successes. In the future, modification of the existing HTLs and ETLs with additives or interfacial materials to obtain balanced transport and improved charge carrier extraction will be beneficial for boosting PCE. To better address the stability and reproducibility problem, highly efficient dopant-free small molecular HTMs, and low-temperature processable inorganic HTLs with high mobility and conductivity, which can eliminate the negative effects of dopants and additives, are greatly desired. Especially, with dopant-free organic HTMs, at least the degradation of perovskite by moisture will be inhibited. Moreover, developing charge-transport materials that can restrict ion migration in perovskite will benefit both PCE and stability. We believe that with synergetic efforts in developing high-performance light-harvesting layers and charge-transport layers, PSCs could eventually be a complementary power source in industry and our daily life.

Conflict of interest

The authors declare that they have no conflict of interest.

Acknowledgments

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Appendix A. Supplementary materials

Supplementary materials to this article can be found online at <https://doi.org/10.1016/j.scib.2020.04.021>.

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