Constructing Molecular Bridge for High-Efficiency and Stable Perovskite Solar Cells based on P3HT Hole Transport Material

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Article

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Abstract

Poly (3-hexylthiophene) (P3HT) is one of the most attracting hole transport materials (HTMs) for the pursuing of stable, low-cost and high-efficiency perovskite solar cells (PSCs). However, the poor contact and the severe recombination at P3HT/perovskite interface lead to a low power conversion efficiency (PCE). Thus, we have constructed a molecular bridge, MDN, whose malononitrile group can anchor the perovskite surface while triphenylamine group can form \( \pi - \pi \) stacking with P3HT, to form a charge transport channel. In addition, MDN was also found effectively passivate the defects and reduce the recombination to a large extent. Finally, a PCE of 22.87\% has been achieved with MDN doped P3HT (M-P3HT) as HTM, much higher than the efficiency of PSCs with pristine P3HT. Furthermore, MDN gave the un-encapsulated device an enhanced long-term stability that 92\% of its initial efficiency has been maintained even after two months of aging at 75\% relative humidity (RH) followed by one month of aging at 85\% RH in the atmosphere, and the PCE has not been changed after operating at the maximum power point (MPP) under 1 sun illumination (~45 °C in N\(_2\)) over 500 hours.

Introduction

Organic–inorganic hybrid perovskite solar cells (PSCs) with the power conversion efficiency (PCE) over 25\%\(^1\)–\(^4\) is widely considered as one of the most promising photovoltaic technologies for its low-cost manufacturing process and appealing photovoltaic performances. In the typical n–i–p type PSCs, hole transport materials (HTMs) are necessary that they can extract and transport holes to enhance the efficiency, and simultaneously block the moisture ingestion so as to be the last barrier of perovskite degradation. The doped 2,2′,7,7′-tetrakis[N,N-di(4methoxyphenyl)amino]-9,9′-spirobifluorene (Spiro-OMeTAD) with appropriate hole mobility and energetic level alignment is the most commonly used HTM\(^5\)–\(^8\). However, the achievement of high efficiency is always accompanied with the substantial instability arisen from the hygroscopicity and fluidity of dopants that can lead to perovskite corrosion and dopant migration\(^9\)–\(^13\), which is detrimental to the long-term stability and commercialization of PSCs\(^14\)–\(^18\).

The development of dopant-free HTMs is thus viewed as an important solution. Organic conjugated materials with sophisticated structures thus have been designed and synthesized as dopant-free HTMs for efficient and stable PSCs. Those include small molecular systems, such as D − A − \( \pi \) − A − D-type DTP-C6Th\(^19\), 1,10-phenanthroline (YZ22)\(^20\), DTB-FL, Ni phthalocyanine (NiPc)\(^21\), etc, and polymeric systems, such as 2DP-TDB\(^22\), phenanthrocarbazole 6 (PC6)\(^23\), Mes-TABT\(^24\), etc. Interestingly, the common characteristic of all these materials is the claimed PCE, as high as over 21\%, but none of them can really substitute the doped Spiro-OMeTAD, even in lab scale. We conjecture this dilemma is caused by their delicate molecular structures, leading to the cumbersome synthesis and high cost, and their poor reproducibility on performances in PSCs due to the identical treatment. In addition, Zhang et al. reported a high PCE, ca. 20\%, based on the undoped Spiro-OMeTAD through solvent-annealing assisted thermal evaporation (SATE)\(^25\). However, this complex evaporation technique is not applicable for most laboratories and the reproducibility still needs verification.
In the meanwhile, utilizing the existing p-type semi-conductors with high hole mobility and mature fabrication process, for example, poly (3-hexylthiophene) (P3HT), as HTMs is another significant attempt. Nevertheless, P3HT adopts the “edge-on” stacking arrangement, that is the alkyl side chains directly contact perovskite film, resulting in an electronically poor contact at perovskite/P3HT interface, which can aggravate the non-radiative recombination loss of PSCs. Thus, the pristine P3HT as HTMs can only achieve a PCE as low as 16%.

In order to solve this problem, interfacial engineering is widely applied. For example, implementing BTCIC-4Cl or copper(I) thiocyanate (CuSCN) at perovskite/P3HT interface to passivate the surface defects of perovskite can give a reported efficiency of ~ 16% for CsPbI₂Br-based devices. Seo et al. introduced a layer of n-hexyl trimethyl ammonium bromide (HTAB) to modulate the packing of P3HT and obtained a PCE over 22% for (FAPbI₃)₀.₉₅(MAPbBr₃)₀.₀₅-based device. And Sun et al. further doped P3HT with gallium(III) acetylacetonate (Ga(acac)₃) to reduce the interfacial recombination loss and demonstrated an enhanced PCE over 24%. While, regarding to the hole transport process, Hu group had improved the hole mobility by directly changing the “edge-on” packing of P3HT into “face-on” packing via SMe-TATPyr doping engineering. Clearly, researchers have taken various strategies to ameliorate the poor performance of P3HT in PSCs, whereas the poor contact issue at perovskite/P3HT interface still has not been stressed.

Herein, we have introduced a molecular bridge (MDN) to electronically link perovskite films with P3HT. In this MDN bridge, the malononitrile group anchors perovskite and passivates its surface defects, while triphenylamine (TPA) group is to form π − π stacking with the segments of P3HT. Finally, in PSCs, a PCE of 22.87% has been achieved with an enhanced open-circuit voltage (V_OC), from 0.88 ± 0.03 to 1.15 ± 0.02, and fill factor (FF), from 52.91% ± 5.14–75.02% ± 3.09%. Moreover, the device has remained more than 90% of its initial efficiency when ageing at 75% and then 85% RH (25 °C) over 2100 h.

Results And Discussion

Molecular stacking structures

The target small molecules, MDN and RDN, are shown in Fig. 1a, and the detailed synthetic route was given in Scheme S1. Among, RDN was designed because of its similar molecular polarity with MDN to exclude the effect of molecular polarity of MDN on its performance in PSCs. Their optical and electrochemical properties were then examined by UV – vis absorption spectrometer and cyclic voltammetry (CV), where MDN and RDN were determined to have the appropriate energetic levels, ca. the highest occupied molecular orbital (HOMO) of − 5.31 and − 5.26 eV and the lowest unoccupied molecular orbital (LUMO) of − 3.28 and − 3.24 eV, matching well with perovskite (Fig. S1, S2, Table S1).

Further, we added MDN (4 mg) and RDN (4 mg) in P3HT (10 mg·mL⁻¹), separately, denoted as M-P3HT and R-P3HT, and compared their optical and energetic structures. The packing structures of M-P3HT and
R-P3HT on silicon wafer was compared with bare P3HT via grazing-incidence wide-angle X-ray scattering (GIWAXS). From Fig. 1b, c, the obvious in-plane (010) diffraction rings of P3HT, M-P3HT and R-P3HT are located at $q_{xy} = 1.65, 1.62$ and $1.64 \text{ Å}^{-1}$, with the corresponding $\pi - \pi$ stacking distances calculated to be $3.81, 3.88$ and $3.83 \text{ Å}$, respectively. And the out-of-plane (100), (200) and (300) diffraction rings of P3HT, $q_z = 0.387, 0.771$ and $1.166 \text{ Å}^{-1}$, are similar to those of M-P3HT and R-P3HT, both $q_z = 0.387, 0.761$ and $1.138 \text{ Å}^{-1}$ (Fig. 1d). In addition, P3HT, M-P3HT and R-P3HT deposited on perovskite ($\text{Cs}_{0.05}\text{FA}_{0.85}\text{MA}_{0.10}\text{Pb(Br}_{0.03}\text{I}_{0.97})_3$) have given the same stacking arrangement as illustrated in Fig S3. Thus, MDN and RDN barely changed the “edge-on” packing structure of P3HT.

Simultaneously, their optical and electronic properties were compared. As shown in Fig. S4 – S6, through UV – vis and ultraviolet photoelectron spectroscopy (UPS) measurements, no significant difference can be distinguished between P3HT, M-P3HT and R-P3HT. Similarly, the hole mobility was measured by space charge limited current method based on the hole-only device structure of ITO/PEDOT:PSS/HTM/Ag. And the hole mobilities of $3.03 \times 10^{-3} \text{ cm}^2\cdot\text{V}^{-1}\cdot\text{s}^{-1}$ for P3HT and $\sim 2.88 \times 10^{-3} \text{ cm}^2\cdot\text{V}^{-1}\cdot\text{s}^{-1}$ for M-P3HT or R-P3HT were obtained (Fig. 1e). This slightly smaller hole mobility is consistent with the minute expansion of the $\pi - \pi$ stacking distance when MDN and RDN was adopted. Moreover, similar conductivities were observed for P3HT, M-P3HT and R-P3HT films (Fig. S7). Therefore, the addition of MDN and RDN has negligible influence on the physical properties of P3HT.

**Photovoltaic performances**

The photovoltaic performance of M-P3HT and R-P3HT HTMs in PSCs has been examined by fabricating n-i-p type devices with configuration of FTO/SnO$_2$/Cs$_{0.05}$FA$_{0.85}$MA$_{0.10}$Pb(Br$_{0.03}$I$_{0.97}$)$_3$/HTM/Ag, as shown in the cross-sectional scanning electron microscopy (SEM) image (Fig. 2a, Fig. S8). The thickness of each layer was estimated to be 560, 20, 610, 30, and 100 nm, respectively. The top view SEM images in Fig. S9 have clearly demonstrated a smooth and even surface of M-P3HT and R-P3HT as P3HT.

The $J$ – $V$ curves of the champion devices have been plotted in Fig. 2b, that the PCEs of devices based on P3HT, M-P3HT and R-P3HT HTMs are 12.48% ($V_{OC} = 0.92 \text{ V}, J_{SC} = 23.21 \text{ mA}\cdot\text{cm}^{-2}, FF = 58.73\%$), 22.87% ($V_{OC} = 1.16 \text{ V}, J_{SC} = 24.72 \text{ mA}\cdot\text{cm}^{-2}, FF = 80.24\%$) and 16.66% ($V_{OC} = 1.05 \text{ V}, J_{SC} = 23.90 \text{ mA}\cdot\text{cm}^{-2}, FF = 66.54\%$), respectively. Obviously, all the parameters, including $V_{OC}$, $J_{SC}$ and $FF$, have been enhanced in PSCs with M-P3HT, which can be highly related to the efficient charge dynamic process throughout the devices. Additionally, a significantly mitigated hysteresis was observed when adopting M-P3HT as HTM, with its hysteresis index as low as 2.6%, comparing with those of P3HT and R-P3HT, ca. 78.8% and 20.1% (Table S2). The external quantum efficiency (EQE) spectra along with the integrated $J_{SC}$, ca. 22.6, 23.9, 22.9 mA·cm$^{-2}$ for the device with P3HT, M-P3HT, and P-P3HT, further confirm the better performance of M-P3HT (Fig. 2d).

By analyzing the statistics of 20 independent devices prepared at the same condition, the reproducibility of PSCs with P3HT, M-P3HT and R-P3HT has been demonstrated. Figure 2c presents the histogram of the
PCEs, where the average efficiency of P3HT is only 10.73%, much lower than that of R-P3HT (15.33%) and M-P3HT (21.19%). The average $V_{OC}$ and $FF$ show the same trend (Fig. S10, S11, Table S3), agreeing well with the $J$–$V$ curves in Fig. 2b. Figure 2e shows the steady power output (SPO) of PSCs with various HTMs at their maximum power point (MPP) bias over 180 s. The devices with P3HT, M-P3HT and R-P3HT have achieved a stable power output of 10.31%, 22.25% and 14.76%, respectively. The devices were then placed in a nitrogen atmosphere and subjected to light stability test at MPP. As shown in Fig. 2f, the device based on M-P3HT shows the nearly same efficiency as its starting state even after more than 500 hours of light soaking, while the other two kinds of devices present severe efficiency degradation.

**Molecular bridge of MDN**

Based on the above results, it is obvious that MDN plays a significant role in PSCs with P3HT HTM. Hence, we utilized the X-ray photoelectron spectroscopy (XPS) to detect the surface of perovskite. When capped with P3HT or R-P3HT, the binding energies (BEs) of Pb 4f and I 3d barely changed comparing with pristine perovskite film (Fig. S12), which is different from the case with M-P3HT. That in Fig. 3a and 3b, the BEs of Pb 4f and I 3d shift to higher values with 560 meV and 330 meV indicates a prominent change in electronic environment of the perovskite surface. Simultaneously, the metallic Pb$^0$ with BEs at 141.51 and 136.59 eV that arises from the decomposition of PbI$_2$ caused by light and X-ray irradiation has been effectively inhibited by M-P3HT (Fig. 3a). In addition, from the XPS spectra of perovskite, perovskite/P3HT and perovskite/M-P3HT in Fig. 3c and Fig. S13, the binding energies of N 1s peaks based on perovskite/M-P3HT is different from others. By peak splitting, perovskite/M-P3HT shows the characteristic peaks of C = N at 401.53 eV and C – N at 400.36 eV from perovskite films and the peaks of N– (Ph)$_3$ at 402.36 eV and C ≡ N at 398.79 eV ascribing to M-P3HT. More interestingly, this BE of C ≡ N bond is smaller than that of pristine M-P3HT (399.77 eV). Combining with a stronger BE of Pb 4f in perovskite/M-P3HT (Fig. 3a), it is concluded that an electrostatic coupling between the N atoms with high electron cloud density in malononitrile group of MDN and the uncoordinated Pb exists, and therefore, building a charge transportation path between perovskite and M-P3HT.

This charge transportation path is also verified by inserting a thin (to ensure the charge tunneling effect) layer of inert polystyrene (PS) between perovskite and M-P3HT in order to isolate their direct contact. As shown in Fig. S14, S15, Table S4, S5, after PS modification, the device efficiency of PSCs with P3HT HTM increases, and the efficiency keeps constant with R-P3HT HTM, whereas the photovoltaic performance of PSCs with M-P3HT declines significantly, that the average efficiency drops from 21.19–18.35%. Therefore, these results confirm that this electronic coupling built by MDN and perovskite has played a significant role for the photovoltaic performance of PSCs (Fig. S18a).

Further, MDN and RDN are inserted as an interface layer between perovskite and P3HT to build the PSCs. It is found that RDN only improves interfacial contact with PCE increases from 10.73–13.45%, while for MDN, the PCE is 16.30%, much lower than that of the PSC with 22.87% of M-P3HT (Fig. S16, S17, Table
It is clear that as interfacial layer, MDN fails to form a charge transportation channel, which is possibly due to its disordered stacking with P3HT, as shown in Fig. S18b.

DFT calculations were adopted to further investigate the interactions between MDN/RDN, P3HT and perovskite. Fig. S19–22 show the optimized structures of perovskite–molecule (MDN/RDN) dimer complexes, where the electron withdrawing groups, di-cyanovinyl and 3-ethylrhodanine, of MDN or RDN anchor the PbI-terminated perovskite surface, respectively. Figure 4a–d present the charge density differences between perovskite (with and without I vacancies, VI) and MDN/RDN, named as PVK-VI/MDN, PVK/MDN, PVK-VI/RDN and PVK/RDN, respectively. It can be seen that the electron withdrawing groups of MDN/RDN attract the electron from Pb ion in perovskite, while the degree of charge transfer in MDN-dimer complexes is greater than that in RDN-one. Meanwhile, the Pb ion in perovskite with I vacancies has a more electron depletion than that without I vacancies. And the lowest binding energy ($\Delta E$) can be obtained in PVK-VI/MDN, indicating their strongest interaction to let MDN anchor on perovskite surface.

While on the other side, the interaction between MDN and P3HT was analyzed by DFT calculations. From the localized orbital locator (LOL) in Fig. S23, MDN has shown a thorough $\pi$-conjugated structure. And in Fig. 4e and 4f, $\pi-\pi$ stacking of MDN and P3HT can be observed via the analysis of independent gradient model based on Hirshfeld partition (IGMH). In particular, we find that triphenylamine group of MDN has a greater degree of $\pi-\pi$ stacking and stronger interaction with P3HT when comparing the geometries in Fig. 4e and 4f.

To conclude, despite RDN and MDN have the similar polarity and optical and electronic properties, MDN can not only anchor the surface of perovskite, but also $\pi-\pi$ stack with P3HT, forming a charge transportation channel as a molecular bridge. This explains the fact that when MDN was merely deposited as the interfacial layer between perovskite and P3HT, in the PSCs, its PCE is not as high as that based on M-P3HT due to its poor $\pi-\pi$ stacking with P3HT.

Further, we have checked D – D–A molecular structure of MDN by design and synthesis of D – A and $\pi–\pi$ molecules, PDN and TDN (Fig. S24). After utilizing PDN and TDN in PSCs with the same procedure as MDN, it is found that they could not get the similar effect as MDN (Fig. S25, S26, Table S8, S9), suggesting the unique of the MDN.

**Defect passivation and stability**

On account of the interaction between MDN and perovskite, the defect density of perovskite was estimated with hole-only devices by space-charge limited current (SCLC) method. The defect densities ($N_t$) can be calculated from the trap-filled limit voltage ($V_{TFL}$) in Fig. 5a, corresponding to $1.39 \times 10^{16} \text{cm}^{-3}$, $9.21 \times 10^{14} \text{cm}^{-3}$, and $6.35 \times 10^{15} \text{cm}^{-3}$ for the devices with P3HT, M-P3HT, and R-P3HT, respectively. The obtained ultra-low trap density of device with M-P3HT, only 1/15 of that with P3HT, explains the minimal non-radiative recombination. Besides, their hole mobility was calculated from the curves in Child region (Fig. S27), ca. $9.86 \times 10^{-6} \text{cm}^{2} \cdot \text{V}^{-1} \cdot \text{s}^{-1}$, $4.10 \times 10^{-4} \text{cm}^{2} \cdot \text{V}^{-1} \cdot \text{s}^{-1}$, $1.20 \times 10^{-5} \text{cm}^{2} \cdot \text{V}^{-1} \cdot \text{s}^{-1}$, respectively, further confirming the MDN molecular bridge.
Electrical impedance spectroscopy (EIS) has been applied to assess the carrier transport dynamics. The Nyquist plot in Fig. 5b shows that M-P3HT HTM gave PSCs a smaller transfer resistance ($R_{tr}$) and a larger recombination resistance ($R_{rec}$). Besides, the capacitance – frequency plots (Fig. 5c) present a 2 orders of magnitude decrease of capacitance at low frequency for devices with M-P3HT or R-P3HT when comparing with conventional PSC based on P3HT HTM, owing to the fact that MDN brings about an effective defect passivation along with the formation of charge transfer channels, resulting in reducing the charge accumulation at perovskite/HTM interface$^{25,40}$.

The voltage dependence on light intensity was plotted in Fig. 5d to investigate the charge recombination behavior in the PSCs. It is clear that M-P3HT based devices exhibit higher $V_{OC}$ values than the devices based on P3HT and R-P3HT, indicative of the less recombination during the photovoltaic process$^{41,42}$. Moreover, the smaller linear slope of PSC with M-P3HT (1.32 kT·q$^{-1}$) than that with R-P3HT (1.61 kT·q$^{-1}$) and P3HT (1.73 kT·q$^{-1}$) further verified this argument.

While the leakage current density obtained from $J - V$ curves measured in dark is much lower in device with M-P3HT, its saturation current density ($J_0 = 9.7 \times 10^{-9}$ mA·cm$^{-2}$) is also 2 orders of magnitude lower comparing with PSCs with P3HT and R-P3HT ($J_0 = 3.6 \times 10^{-7}$ mA·cm$^{-2}$, $J_0 = 1.5 \times 10^{-7}$ mA·cm$^{-2}$), as shown in Fig. 5e. And the current density of M-P3HT device increases significantly in the high voltage region (0.8 – 1.5 V), indicating that the perovskite/M-P3HT interface gives a better charge transportation$^{43,44}$.

At last, to investigate the influence of HTM on device’s long-term stability at moisture atmosphere, we have monitored the PCE of PSCs with various HTMs without any encapsulation at a high relative humidity gradient (75% RH for 60 days and then 85% RH for 30 days) at 25 °C in atmosphere. As shown in Fig. 5f, the efficiency of M-P3HT and R-P3HT based devices hardly changed during the first two months, which is in sharp contrast with the situation for P3HT based devices, that only 50% of the initial efficiency was maintained. More importantly, further increasing the relative humidity to 85%, with the protection of M-P3HT and R-P3HT, their PSCs still exhibited ~ 92% and ~ 80% of their initial efficiency after another one month aging, showing an effective moisture blockage of M-P3HT. This enhanced stability is considered coming from the high hydrophobicity of M-P3HT (Fig. S28), the tighter molecular structure and the reduction of perovskite surface defects which acts as the starting sites for perovskite decomposition.

**Conclusion**

In summary, we have constructed a molecular bridge, MDN, to improve the poor contact between perovskite and P3HT, where the malononitrile group of MDN anchors the surface of perovskite and the triphenylamine group forms a tight π – π stacking with the conjugated polythiophene segment in P3HT. And thanks to this charge transportation channel, the fabricated PSCs have achieved a PCE as high as 22.87% with negligible hysteresis. Moreover, the un-encapsulated PSCs have shown a long-term stability
at a high relative humidity in atmosphere, that more than 90% of the efficiency has been maintained over 3 months aging, as well as a light stability over 500 h at MPP. These distinguishes results clearly show a bright future for the industrialization of low-cost, stable and efficient PSCs by utilizing the commercial P3HT-based HTMs.

**Declarations**

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**Author contributions**

Dongdong Xu, Zhiming Gong, Yue Jiang and Jinwei Gao conducted the idea. Zhiming Gong designed and synthesized target molecules. Dongdong Xu performed the electro-optical characterizations and the experiments on solar cells. Yancong Feng performed the DFT calculation. Zhen Wang coordinated the research activity. Xingsen Gao, Xubing Lu, Guofu Zhou and Jun-Ming Liu contributed to the revision of the manuscript. The manuscript was written by Dongdong Xu, Yue Jiang and Jinwei Gao. Yue Jiang and Jinwei Gao directed the work. All the authors approved the final version of the manuscript.

**Competing interests**

The authors declare no competing interests.

**References**


**Figures**
Figure 1

Molecular packing characterization of P3HT, M-P3HT and R-P3HT. (a) Molecular structure of MDN, RDN and P3HT. (b) 2D GIWAXS patterns. (c) 1D intensity profiles of relevant films along the in-plane and (d) out-of-plane directions. (e) Hole mobility measurement characteristics by the space charge limited current (SCLC) method.
Figure 2

PSCs Performances. (a) Cross-sectional SEM image of PSC with M-P3HT. (b) J–V curves of the champion devices based on P3HT, M-P3HT and R-P3HT, respectively. (c) PCE histogram obtained from 20 PSCs. (d) EQE spectra and integrated photocurrent curves of the device with P3HT, M-P3HT, and R-P3HT. (e) The steady-state PCE. (f) The devices stability at maximum power point (MPP) under full solar illumination (AM 1.5G, 100 mW·cm⁻²) in glovebox.
Figure 3

**Molecular bridge characterization of M-P3HT.** High-resolution XPS spectra of (a) Pb 4f spectra, (b) I 3d spectra, and (c) N 1s spectra.
Figure 4

Snapshots of molecular bridge. Charge density differences of (a) PVK/MDN, (b) PVK-V\textsubscript{i}/MDN, (c) PVK/RDN and (d) PVK-V\textsubscript{i}/RDN. PVK-V\textsubscript{i} denotes the perovskite with I vacancy, and expresses the binding energy. Local perovskite is displayed for clarity. The yellow and cyan regions represent electron accumulation and depletion, respectively. Independent gradient model based on Hirshfeld partition (IGMH) of MDN/P3HT dimer complexes with different geometries (e) and (f).
Figure 5

Passivation of perovskite surface defect and stability of devices. (a) I-V curves of the hole-only devices for quantitative measurement of the trap densities. (b) Nyquist plots of PSCs under the dark condition at 0.8 V bias. (c) Capacitance–frequency plots of PSC devices employing different HTM. (d) The dependence of $V_{OC}$ on light intensity of relevant PSCs. (e) Dark $J–V$ curves of the devices. (f) Stability of the devices in high humidity gradient environment without encapsulation. The error bars represent the standard deviations from ten samples for each condition.

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