



Full paper

Boosting performance of inverted organic solar cells by using a planar coronene based electron-transporting layer



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ABSTRACT

In this work, an alcohol-soluble, low-temperature processable and relatively thickness insensitive electron-transporting layer (ETL) comprising a planar coronene derivative, CDIN, was exploited to effectively enhance the photovoltaic performance of various inverted organic photovoltaics (OPVs). Besides the decent charge-transporting property, such CDIN ETL was manifested to facilitate the face-on orientation of atop bulk-heterojunction (BHJ) layers as evidenced by GIWAXS analysis, which might benefit from its discotic geometry endowed with strong face-on π - π stacking in solid-states and better compatibility to the constituent organic photoactive components. Consequently, an enhancement of over 9% in PCE can be achieved in the state-of-the-art fullerene-based OPVs to yield a PCE of 11.2% while over 13% enhancement can be realized in the representative non-fullerene OPVs to yield a PCE of 9%.

1. Introduction

Organic photovoltaics (OPVs) have drawn significant attention due to their potential for low cost, light weight, printable and high-throughput solution processing [1–3]. In the past decades, tremendous progress had been made in improving the performance of OPVs and a promising power conversion efficiencies (PCEs) of over 12% has been recently demonstrated [4–10]. The stratified OPVs can be generally classified into two types, conventional and inverted architecture, according to the sequence of the employed charge-transporting layers (CTLs). Among the exploited configurations, the inverted configuration attracts more widespread research interests due to its better light-harvesting capability and stability [11–15].

Currently, one urgent issue in the OPV development is to increase its compatibility to roll-to-roll (R2R) printing techniques. This necessitates the development of OPVs that comprises thick constituent layers, including photoactive layer and CTLs. Hence, significant attention has been paid to develop new materials with high charge mobility or conductivity for both BHJ layers and CTLs to afford better device performance under a thicker condition [16–18]. For example, for the

photoactive layers, several groups have lately developed semi-crystalline polymers endowed with respectable charge mobility as a result of the optimized intermolecular interactions to yield decent PCEs using thick BHJ films (~ 300–1000 nm) [19–23].

Similar principle has also been applied when seeking efficient and thickness-insensitive CTLs, particular for the electron-transporting layers (ETLs). Note that the exploitation of proper CTLs and interfacial optimization also plays a pivotal role to promote the performance advancement [6,24–31]. Such interface engineering not only improves the charge-transporting of the derived devices but also can impact the film evolution of the bulk-heterojunction (BHJ) layer grown on top of them since it is critically correlated with the surface energy and surface texture of the seeding layers [32–36].

Thus far, zinc oxide (ZnO) is the most commonly used ETL due to its high electron mobility, suitable energy levels, and high transparency across the ultraviolet-visible spectral range [13]. However, several deficiencies of ZnO ETL have been gradually identified, such as the defective surface that acts as charge trapping sites and unsatisfactory compatibility to the organic BHJ layers [37,38]. In this regard, exploitation of efficient organic ETLs has recently attracted substantial

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research attention and various compounds including fullerene-, poly [(9,9-bis(3'-(*N,N*-dimethylamino)propyl)-2,7-fluorene)-*alt*-2,7-(9,9-dioctylfluorene)] (PFN)-, and perylene-diimide (PDI)-based derivatives has been successfully developed and applied to deliver high PCEs [3,16,18,26,36,39–43]. However, most high-performance OPVs reported to date still employed very thin organic CTLs with an optimal thickness around 5–10 nm due to their limited charge mobility. On the other hand, the increase of these organic ETLs' thickness might incur the parasitic absorption in the visible light region, especially for the inverted OPVs wherein the ETL is located at the light-incoming side.

We have lately developed a low-temperature, alcohol solution-processable organic ETL consisting of an amine functionalized discotic coronene diimide (CDIN) that can successfully endow the derived perovskite solar cells with high efficiency [44]. Besides the advantages of low-temperature, alcohol solution-processability of CDIN, the discotic geometry of coronene diimide core with an expanded π -conjugated planar structure endows an efficient face-on π - π stacking in the solid-states, which is beneficial for providing respectable charge carrier mobility in vertical direction. Further, the weak intensity and large band-gap of CDIN can reduce the parasitic absorption efficiently in the visible light region for the inverted OPVs. We herein systematically explore its efficacy in various inverted OPVs including fullerene and non-fullerene BHJ systems. By using the CDIN as the bottom ETL, all the studied devices exhibited an improved PCE compared to the control device using a typical ZnO ETL. The underlying mechanism for the PCE enhancement has been systematically studied. On one hand, such planar discotic CDIN possesses a favorable face-on π - π stacking in solid-states to afford respectable charge carrier mobility. On the other hand, it bears suitable surface energy and surface texture to effectively modulate the atop BHJ morphology, resulting in a more optimized crystal formation and orientation.

Consequently, an enhancement of over 9% in PCE can be achieved in the state-of-the-art fullerene-based OPVs to yield a PCE of 11.2% while over 13% enhancement can be realized in the representative non-fullerene OPVs to yield a PCE of 9%. Moreover, benefitting from its superior transparency and mobility, the CDIN ETL shows a relatively thickness insensitive feature for the derived OPVs, which can still afford respectable performance at a relatively thick thickness of 25 nm. More intriguingly, the CDIN ETL can enable the use of a thicker BHJ layer (~380 nm) since it can positively impact the BHJ evolution to result in

a more optimized morphology.

2. Result and discussion

Fig. 1a presents the molecular structures of the compounds that constitute the studied BHJ systems in this study and the CDIN ETL, wherein the studied BHJ system includes the blends of PTB7-Th/PC₇₁BM, r-PTB7-Th [45]/PC₇₁BM, PTB7-Th/ITIC, and r-PTB7-Th/ITIC. As noted in our previous work, [44] the CDIN ETL can be simply processed by methanol and shows a well solvent resistance to the common non-polar solvents (Fig. S1), like chlorobenzene and *o*-dichlorobenzene (DCB), that are generally used to process BHJ layers. This nature enables the CDIN ETL to serve as a robust bottom ETL in the inverted OPVs. The corresponding frontier energy levels of these compounds measured by cyclic voltammetry (CV) are summarized in Fig. 1b and the fabricated device configuration (ITO/ETL/BHJ/MoO₃/Ag) is illustrated in Fig. 1c, for which three different ETLs are investigated herein, including regular ZnO, ZnO/CDIN, and CDIN.

The current density-voltage (J - V) characteristics of the fabricated inverted OPVs under 1-sun illumination are presented in Fig. 2a–d and the corresponding photovoltaic parameters like open-circuit voltage (V_{OC}), short-circuit current (J_{SC}), and fill factor (FF) are summarized in Table 1. The corresponding external quantum efficiency (EQE) of these fabricated OPVs are illustrated in Fig. 2e–f. As shown, the integrated J_{SC} s from the EQE spectrum are in good agreement with the values obtained in the J - V curves (within 3% mismatch), validating the accuracy of our measurements and the results.

All the studied devices using the regular ZnO ETL showed decent PCEs of 9.30%, 10.14%, 7.28%, and 7.97% for the PTB7-Th/PC₇₁BM, r-PTB7-Th/PC₇₁BM, PTB7-Th/ITIC, and r-PTB7-Th/ITIC BHJs, respectively. Very encouragingly, when using a CDIN ETL to modify or replace the original ZnO ETL, all the studied devices yielded enhanced PCEs. As can be seen, the PCE_{max} of the r-PTB7-Th/PC₇₁BM device was improved from 10.14% (ZnO ETL), 10.62% (CDIN ETL; J_{SC} = 17.66 mA/cm², V_{OC} = 0.83 V, FF = 0.73), to 11.17% (ZnO/CDIN ETL; J_{SC} = 17.91 mA/cm², V_{OC} = 0.83 V, FF = 0.75). This improvement mainly results from the enhanced short-circuit current (J_{SC}) and fill factor (FF) and similar phenomenon was also observed in the other studied BHJ systems. For the r-PTB7-Th/ITIC BHJ system, the device using a CDIN ETL showed a higher PCE_{max} of 8.95% (J_{SC} = 16.33 mA/cm², V_{OC} = 0.84 V, FF =

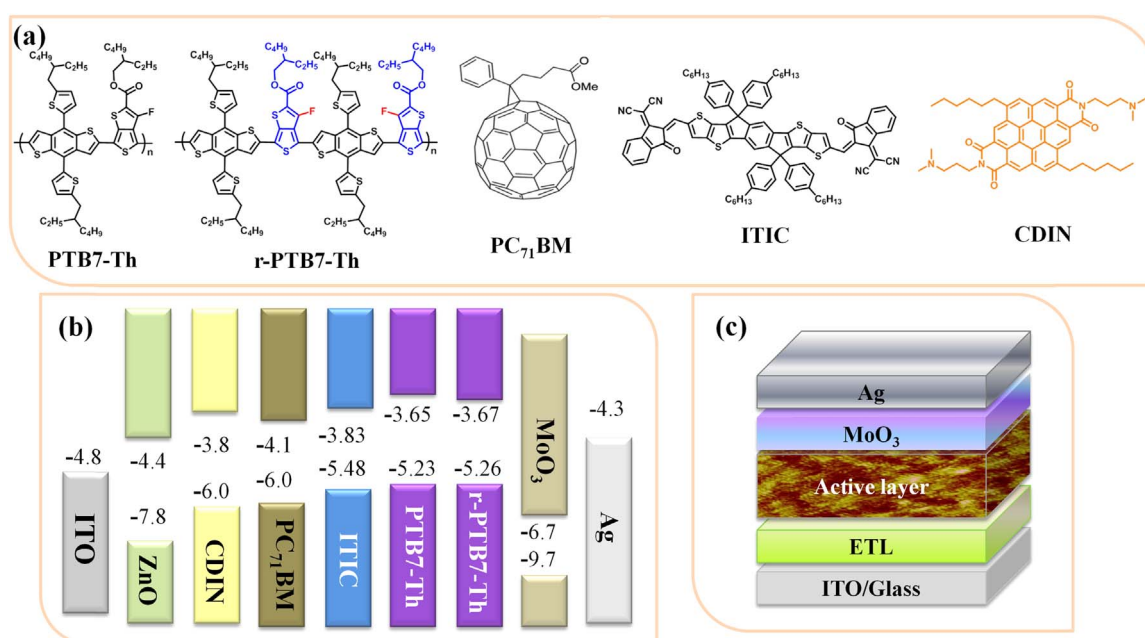


Fig. 1. (a) The chemical structures, (b) the energy levels diagram of the studied compounds and (c) the schematic device configuration used in this work.

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