Organic Semiconductors for Vacuum-Deposited Planar Heterojunction Solar Cells

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ABSTRACT: Relative to widely used solution-processed bulk heterojunction organic solar cells (OSCs), planar heterojunction (PHJ) OSCs by vacuum-depositing active layers sequentially avoid tedious control of the blend film morphology, and it is easy to understand the physical process at the donor/acceptor interface. Here we summarize the developments of electron donor and acceptor materials for vacuum-deposited PHJ OSCs in the past decades and discuss the relationship between molecular structure and device performance. Finally, the challenges and prospects for the development of vacuum-deposited PHJ OSCs are also proposed. In addition to some basic requirements for high performance organic photovoltaic materials, such as broad and strong absorption, matched energy levels between donors and acceptors, and high charge carrier mobility, we suggest that extending the exciton diffusion length of organic photovoltaic materials should boost PHJ OSCs gradually become another option for organic photovoltaic application.

1. INTRODUCTION

Organic solar cells (OSCs) show considerable potential in commercial applications due to their unique advantages, such as light weight, flexibility, and semitransparency.1 In 1986, the first planar heterojunction (PHJ) organic photovoltaic device was fabricated by vacuum evaporation with an efficiency of about 1%.2 Since then, the vacuum-deposited PHJ structures were extensively researched. Unfortunately, the exciton diffusion distance of the organic photovoltaic materials was only 5−20 nm, which limited the further improvement of photovoltaic performance.3

The bulk heterojunction (BHJ) solar cells greatly increase the donor/acceptor (D/A) interfacial area compared with the PHJ solar cells, leading to enhanced efficiency of the exciton separation. So far, the mainstream devices are still employing BHJ structures with solution-processed active layers. The power conversion efficiency (PCE) of OSCs has exceeded 18%4 with the emergence of efficient nonfullerene fused ring electron-acceptor materials. A crucial ingredient to the surge of PCE is the morphology of BHJ films which has an important impact on the charge generation, transfer, and collection. An ideal morphology of the BHJ active layer was required to have a bicontinuous interdigitated donor and acceptor network to achieve effective exciton dissociation and electron/hole transport.5 However, the actual morphologies are far from this requirement due to the random mixing nature of the donor and acceptor during solution processing. In order to achieve the optimal morphologies, many strategies have been developed to optimize the morphologies of BHJ films, such as the choice of solvent, additives, thermal annealing, solvent vacuum annealing, and among others. Although these strategies have improved the performance of the devices to some extent, the preparation methods of BHJ films are highly sensitive to the organic photovoltaic material properties with unique self-organizing tendencies. The relationship between the structure of the active layer materials and device performance is not clear, mainly because of the complex structure of BHJs.6 Optimizing the domain size and orientation of the blended films remain a big task for new donors or acceptors for BHJ devices. Moreover, the optimized morphology is usually metastable, leading to a decrease in device stability.7

In contrast to BHJ structures, PHJs have some unique advantages:8 (a) the well-defined interfacial structures may eliminate the influence of BHJ morphology metastability, which is beneficial to improving the stability of devices; (b) in PHJ devices, the carrier generation primarily occurs at the planar heterojunction, and then holes/electrons transport within the donor/electron phases, respectively, which induces an enhanced charge carrier lifetime and reduced charge carrier recombination and then benefits the improvements of short-circuit current
density ($J_{oc}$) and fill factors (FF); in contrast, the complex intermixing of the donor and acceptor phases in BHJ leads to an increased charge carrier recombination; (c) D–A vertical separation through tuning the sequence of D and A deposition could be expected to decrease energy disorder, reduce nonradiative combination, and then decrease energy losses, leading to high $V_{OC}$; for example, in CuPc: C$_{60}$ based PHJ and BHJ devices, the $V_{OC}$ of the PHJ device is larger than that of BHJ device, which is a result of longer charge carrier lifetimes and smaller active recombination region; (d) donor and acceptor layers in PHJ can be optimized separately for excellent performance; (e) PHJ structures are employed in many theoretical models for accurately understanding the relationship between molecular structure and device performance, because fewer assumptions and parameters were required.

In this mini-review, we focus on the representative photovoltaic materials in vacuum-deposited PHJ devices. The structures of the vacuum-processed photovoltaic materials used in this review are shown in Figure 1, and the related device data are summarized in Table 1. The current challenges

![Figure 1. Molecular structures of donor and acceptor materials through vacuum deposition.](image1)

![Table 1. Photovoltaic Properties of Vacuum-Deposited PHJ OSCs, Tested under the Illumination of AM1.5, 100 mW cm$^{-2}$, unless Stated Otherwise](table1)

| donor | acceptor | $V_{OC}$ (V) | $J_{SC}$ (mA cm$^{-2}$) | FF (%) | PCE (%) | ref |
|-------|----------|--------------|--------------------------|--------|---------|-----|
| CuPc  | PTCBI    | 0.46         | 2.6                      | 65     | 0.95$^a$ | 2   |
| SubPc | C$_{60}$ | 0.97         | 3.36                     | 57     | 2.1     | 10  |
| SubNc | C$_{60}$ | 0.79         | 6.1                      | 49     | 2.5     | 11  |
| CIAIPc| C$_{60}$ | 0.68         | 6.2                      | 50     | 2.1     | 12  |
| PdPc  | PTCBI    | 0.52         | 4.0                      | 64     | 1.3     | 13  |
| SQ    | C$_{60}$ | 0.75         | 7.13                     | 60     | 3.2     | 14  |
| pentacene | C$_{60}$ | 0.36     | 15                      | 50     | 2.7     | 15  |
| tetracene | C$_{60}$ | 0.58   | 7                       | 57     | 2.3     | 16  |
| tetracene-M | C$_{60}$ | 1.06    | 1.48                     | 34     | 0.5     | 17  |
| tetracene-D | C$_{60}$ | 0.31    | 1.73                     | 37     | 0.2     | 17  |
| DCVST | C$_{60}$ | 0.98         | 10.6                     | 49     | 3.4$^b$ | 18  |
| BTDA-TFA-Bu | C$_{60}$ | 1.17    | 4.04                     | 33     | 1.56    | 19  |
| TPA-2DCN | C$_{60}$ | 0.89    | 3.65                     | 36     | 1.17    | 20  |
| CuPc  | C$_{60}$ | 0.58         | 18.8                     | 52     | 3.6$^c$ | 21  |
| $\alpha$-6T | SubPc  | 1.09  | 7.46                     | 58     | 4.69    | 22  |
| $\alpha$-6T | SubNc | 0.94   | 12.04                    | 54     | 6.02    | 22  |
| SubPc | Cl$_6$SubPc | 1.31 | 3.53                     | 58     | 2.68    | 23  |

$^a$Tested under 75 mW cm$^{-2}$ simulated AM1.5G illumination $^b$Tested under 118 mW cm$^{-2}$ simulated AM1.5G illumination $^c$Tested under 150 mW cm$^{-2}$ simulated AM1.5G illumination
and prospects of vacuum-deposited PHJ OSCs are also discussed.

2. VACUUM-DEPOSITED ORGANIC PHOTOVOLTAIC MATERIALS

Vacuum deposition techniques have been successfully applied to the organic light-emitting diode (OLED) industry; thus OSCs, prepared through vacuum evaporation are another promising candidate for industrial production, in addition to solution process fabrication like the roll to roll method. The key factor to affect the PCE of vacuum-deposited PHJ solar cells is the optical and electrical performance of the small-molecule active layer (donor and acceptor) materials, which possess defined molecular structure, high purity (purified by sublimation), suitability for large-scale production, and good batch repeatability.

2.1. Electron Donor Materials. Since the first PHJ solar cells achieved a PCE of 0.95% employing copper phthalocyanine (CuPc) as donor and perylene tetracarboxylic derivative (PV) as acceptor (Figure 2), researches have focused on enhancing the efficiency of PHJ OSCs through exploring new photovoltaic materials. Dyes, fused acenes, as well as oligothiophenes are considered to be potential donor materials in PHJ OSCs. Dyes, like metallophthalocyanines (MPcs), subphthalocyanine (SubPc), and their derivatives, exhibit high extinction coefficient, high hole mobility, as well as chemical and thermal stability. Thompson and co-workers reported a novel donor, SubPc, which showed stronger absorption and deeper highest occupied molecular orbital (HOMO) level than CuPc. The devices based on SubPc/C60 delivered a PCE of 2.1% with a high open-circuit voltage (VOC) of 0.97 V and JSC of 3.36 mA cm−2. However, the optical absorption of SubPc is still limited to below 600 nm, which is not conducive to increase JSC. Chloroboron-(III) subnaphthalocyanine (SubNc) exhibits a red-shifted absorption (730 nm) relative to that of SubPc, due to the extended conjugation (adding a ring system to the isodole units). When combining C60 acceptor, a PCE of 2.5% was obtained with JSC of 6.1 mA cm−2 in the solar cells employing a SubNc donor. Chloroaluminum phthalocyanine (ClAlPc) possesses an absorption peak at 755 nm, which red-shifted about 135 nm compared to that of CuPc. A PCE of 2.1 ± 0.1% was achieved in PHJ solar cells with depositing ClAlPc and C60 sequentially. In PHJ OSCs, a key limiting factor is short exciton diffusion lengths in photovoltaic materials. In 2009, Jabbour et al. developed a new donor, PdPc, by substituting the Cu atom of CuPc with Pd, and the device demonstrated a higher PCE of 1.3%. The higher performance is attributed to that the heavy metal Pd can provide a rapid conversion route of singlet into the triplet excitons, leading to longer exciton diffusion length (10.1 nm) than that of CuPc (5.8 nm), which was estimated from the best fitting of external quantum efficiency (EQE). Squaraine dye has been effectively applied to PHJ OSCs due to its broad absorption and high extinction coefficients. The devices that used 2,4-bis-[4-(N,N-disobutyramino)-2,6-dihydroxyphenyl] squaraine (SQ) as donor and C60 as acceptor exhibited a PCE of 3.2% under 1 sun and AM1.5G-simulated solar irradiation.

In addition to traditional dyes, fused acenes, such as pentacene and tetracene, have attracted considerable attention due to their extremely high hole mobility. Kippelen and co-workers developed PHJ devices, and employing pentacene as donor and C60 as acceptor achieved a PCE of 2.7% with high EQE values in the red region of the spectrum, which can be attributed to the large exciton diffusion length (65 ± 16 nm) of pentacene film estimated by combining EQE measure and steady-state diffusion equations. In 2005, the PHJ structures were fabricated with tetracene (high mobility and large photosensitivity)/C60, which delivered a PCE of 2.3 ± 0.5% with higher VOC of 0.58 V than that (0.36 V) of the control devices based on the pentacene donor. Thompson et al. reported a new silylthethyl-substituted tetracene monomer (tetracene-M) and dimer (tetracene-D), and the PCE of PHJ devices based on tetracene-M/C60 was 0.5% with a VOC of 1.06 V. The PCE based on tetracene-D was only 0.2%, which is mainly attributed to higher HOMO energy level of tetracene-D (−5.16 eV) than that (−5.36 eV) of tetracene-M.

Thiophenes are potential skeletons for building photovoltaic donor materials due to their good charge-transporting property and strong electron-donating properties. The optical bandgap of unsubstituted quinquephenylene reached 2.5 eV, which cannot capture photons at the wavelength over 500 nm. Forming D−A conjugated structure by introducing electron-withdrawing groups can enlarge the light-absorbing regions and drive the self-organization of oligomers to form an ordered structure. Dicyanovinylene (DCV), which is a strong electron-withdrawing unit, exhibits efficient intramolecular charge transfer when coupled with oligothiophene. Bäuerle and co-workers synthesized a new donor (DCVST), comprised with five α-conjugated thiophene rings (ST) as the electron-donating unit and two DCV groups as the electron-withdrawing unit. DCVST possesses relatively small optical band gap of 1.77 eV and low HOMO energy level (−5.6 eV); thus, an efficient PHJ OSC with PCE up to 3.4% can be achieved under illumination at 118 mW cm−2 simulated sunlight. After that, they developed a novel A−D−A−D−A-type donor (BTDA-TFA-Bu) bearing benzo[c]-[1,2,5]thiadiazole (BTDA) as the core and trifluoroacetyl

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**Figure 2.** Configuration and current–voltage characteristics of an ITO/CuPc/PV/Ag cell. Copyright 1986, AIP Publishing.
(TFA) as the end groups. The vacuum-processed PHJ device based on BTDA-TFA-Bu/C60 delivered a PCE of 1.56% with a high VOC of 1.17 V.\textsuperscript{19} Different from the linear molecules shown above, TPA-2DCN was a star-shaped three-dimensional molecule based on a triphenylamine core derivatized with electron-withdrawing dicyanovinyl groups. TPA-2DCN/C60-based OSCs showed a PCE of 1.17%.\textsuperscript{20}

### 2.2. Electron Acceptor Materials

The acceptor materials play an equally important role with donor materials for high performance vacuum deposition PHJ OSCs. However, there are very few reports in the literature on new acceptor materials for PHJ OSCs. In 2001, Forrest’s group fabricated a device based on CuPc as donor and C60 as acceptor, which delivered a PCE of (3.6 ± 0.2)% under AM1.5 spectral illumination of 150 mW/cm\textsuperscript{2}. Relative to Tang’s device based on CuPc/PTCBI in 1986, C60 shows longer exciton diffusion length, (77 ± 10) Å, compared to that of PTCBI, (30 ± 3) Å, which was estimated by combining the photocurrent test with the theoretical model.\textsuperscript{21} Along with relatively long exciton diffusion length, the anisotropy of electron transfer and good electron transporting made C60 dominate the acceptor materials in vacuum-processed devices.\textsuperscript{22} However, the poor absorption in the visible and deeper lowest unoccupied molecular orbital (LUMO) energy level of C60 limit the photocurrent generation and VOC in C60-based devices, respectively. SubPc not only showed good performance as a donor material but also acted as an electron acceptor when combining some other donors with high energy levels. When employing α-sexithiophene (α-6T) as donor, the device achieved a PCE of 4.7% and 6.0% used SubPc or SubNc as an acceptor due to complementary absorption (Figure 3a).

![Figure 3](image)

**Figure 3.** (a) The absorption spectra of the three active materials complement each other to effectively harvest solar light. (b) Energy-level diagram of the active layers illustrating the two-step exciton dissociation mechanism. (c) The measured EQE (solid lines) and IQE (dashed lines) spectra show efficient photocurrent generation by all three absorbing materials. (d) J–V curves of different active layers.\textsuperscript{23} Copyright 2014, Nature Publishing Group.

and matched energy level (Figure 3b) between α-6T and SubPc or SubNc. Furthermore, a trilayer cascade structure was prepared comprising SubPc and SubNc as acceptors and α-6T as donor, with an efficient two-step exciton-dissociation mechanism. Excitons generated in the SubPc are transferred by long-range Förster energy transfer to the SubNc and subsequently dissociate at the α-6T/SubNc interface. The photocurrent originates from all three photovoltaic materials, resulting in quantum efficiency above 75% between 400 and 720 nm (Figure 3c). In addition, a high VOC of 0.96 V is realized due to the enhanced energy level alignment between α-6T and SubPc/SubNC, leading to a remarkable PCE up to 8.4% (Figure 3d).\textsuperscript{24} Jones and co-workers further adjusted the energy level of SubPc by introducing chlorine atoms to obtain Cl6-SubPc, the PHJ devices based on SubPc, as the donor delivered a highest VOC of 1.31 V. More importantly, it was demonstrated that the devices had better stability when using Cl6-SubPc as an acceptor instead of C60 under continuous illumination in a N\textsubscript{2} atmosphere. The improvement of stability of the SubPc/Cl6-SubPc devices is the result of the lower propensity for photooxidation than that of SubPc/C60 devices.\textsuperscript{25}

### 3. SUMMARY AND OUTLOOK

In this review, some representative organic photovoltaic materials for vacuum-deposited PHJ OSCs were discussed. Several photovoltaic materials, especially donor materials, have been developed, but most of the devices delivered relatively low PCEs. In order to obtain high-performance PHJ OSCs, new high efficiency photoactive materials for PHJ devices should be developed with the following requirements: (a) relatively low molecular weight and excellent thermal stability are preferred requirements for easy sublimation; (b) narrow optical bandgap and high extinction coefficient for obtaining higher \(J_{SC}\); (c) matched HOMO/LUMO energy levels between donor and acceptor to achieve large \(V_{OC}\) and reduce energy loss; (d) long exciton diffusion length for excitons reach the D/A interface effectively and make it possible to prepare relatively thick devices to fully utilize solar energy; (e) high and balanced electron and hole mobility of acceptor or donor, respectively, for higher \(J_{SC}\) and FF.

So far, BHJ OSCs have achieved a PCE of 18.2%\textsuperscript{26} through the fast development of active layer materials and device optimization. There are many molecular design strategies for BHJ OSCs that can be used for reference in designing PHJ photovoltaic materials, such as

1. A—DA’D—A fused-ring conjugation molecule can broaden absorption to the near-infrared region;
2. Asymmetric structure with a high dipole moment leads to strong intermolecular interaction and thus a strong molecular packing for high carrier mobility;
3. Introducing electron-rich or electron-deficient groups adjust the HOMO or LUMO energy level.

Different from photoactive materials for BHJ OSCs, what needs more attention for PHJ devices is that the limited exciton diffusion length of organic materials limits the development of PHJ devices. However, with the development of nonfullerene acceptors (NFAs) for BHJ OSCs, it has been reported that some fused-ring NFAs have longer exciton diffusion distances than those of traditional photovoltaic materials. For example, IDIC (Figure 4a) has high exciton diffusion coefficient of at least 2 × 10\textsuperscript{−2} cm\textsuperscript{2}/s due to local resonant energy transfer.\textsuperscript{25} The rigid planar structure of IDIC contributes to strong self-overlap between the absorption and photoluminescence (PL), which comes from the 0–0 vibronic peak (Figure 4b) and decreased the energetic disorder. The measured energetic disorder (10–23 meV) by the temperature dependence of PL spectra (Figure 4c) is less than thermal energy at room temperature (26 meV), and high exciton diffusion coefficients (Figure 4d) can be maintained even under 80 K. Both strong resonant overlap and low energetic disorder are beneficial to enhancing resonant
energy transfer. The calculated exciton diffusion length of IDIC can achieve 72−87 nm in thin film when applying measured limits for energy disorder of σ = 0−23 meV (Figure 4e), which is much higher than those (5−20 nm) of traditional photovoltaic materials. These gratifying developments on extending exciton diffusion in photovoltaic materials make it possible to achieve high performance PHJ solar cells. We believe vacuum-deposited PHJ OSCs can gradually become another promising option for organic photovoltaic application.

Figure 4. (a) Chemical structure of IDIC. (b) Steady state absorption and photoluminescence spectra of IDIC in both solution and thin film along with their fits to the vibronic progression model. (c) Temperature dependence of 0−0 emission peak energy (extracted from fits to vibronic progression) of neat IDIC film after 680 nm excitation. The range of Gaussian energetic disorder is obtained from the slopes at each extreme (which are noted in the figure) along with the temperature-dependent steady state emission spectra in the inset. (d) Arrhenius plot of exciton−exciton annihilation constants versus reciprocal temperature. (e) Monte Carlo simulations of resonant transport in IDIC thin films. Copyright 2019, American Chemical Society.

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Notes
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