



Cascade organic solar cells with energy-level-matched three photon-harvesting layers

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ABSTRACT

The performances of organic photovoltaic cells were improved by matching the energy levels of three photon-harvesting layers with distinct light absorption spectra. Pentacene, phthalocyanine, and C₇₀, which have shallow, intermediate, and deep energy levels, respectively, were introduced as photon harvesting layers to form a cascade structure and secure appropriate band offsets at all interfaces. This architecture yielded higher values of the open-circuit voltage and short-circuit current density than a standard bilayer structure based on pentacene and C₇₀. A device prepared with metal-free phthalocyanine performed better than a device with Cu phthalocyanine because the energy levels were more appropriately tuned.

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1. Introduction

Recently, organic photovoltaic (OPV) cells have been extensively studied as future energy sources because of their low cost, light weight, and mechanical flexibility [1]. The power conversion efficiencies (PCEs) of OPV cells have been steadily improved by applying several strategies [2–13]. The inherent tradeoff between the light absorption and the exciton diffusion efficiency has hindered improvements in PCE values because optical absorption lengths ($L_A \sim 100$ nm) are typically much longer than exciton diffusion lengths ($L_D \sim 10$ nm) [4,14]. One approach to addressing this problem is to use multilayered photoactive materials with different light absorption spectra. The multilayered structures comprising several photoactive materials can increase the L_A and broaden the span of the solar spectrum that the photoactive layer can absorb, thereby increasing the absorption efficiency. The multilayered structures also do not reduce the efficiency of exciton diffusion by providing multiple junctions where exciton dissociation occurs. Several examples of multilayered structures have been reported including interfacial layer-inserted structures [15–19] and multiple-device stacked structures [10–13]. The device performances of these structures were enhanced by higher open-circuit voltages (V_{OC}); however, the short-circuit current densities (J_{SC}) decreased slightly. The J_{SC} of the interfacial layer-inserted structures was lower than that of simple donor/acceptor bilayer structures due to the reduced charge separation efficiency. In multiple-device stacked structures, the J_{SC} was limited to that of either subcell due to charge carrier recombination at the intermediate electrode. The enhancement of V_{OC} and limitation of J_{SC} in these multilayered

structures are closely related to the energy levels of the layers. However, not many studies have been reported on the control of the energy levels for simultaneous enhancement of both V_{OC} and J_{SC} .

Here, we enhanced both the V_{OC} and J_{SC} of the OPV cells by matching the energy levels of three photon-harvesting layers with different absorption spectra so that the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) levels of the three materials formed a cascade energy band structure. The band offsets at all interfaces in the photoactive layer were designed to be sufficiently large to permit exciton dissociation. To demonstrate this concept, we employed pentacene, phthalocyanine dyes, and C₇₀ as photon-harvesting materials. Pentacene is a p-type material and exhibits a high hole-mobility and shallow HOMO and LUMO levels. Compared to C₆₀, which is a representative n-type material, C₇₀ displays better light absorption properties and deeper HOMO and LUMO levels. The large band offset between the pentacene and C₇₀ could produce an excellent of interlayer material with optimal energy levels. Several phthalocyanine dye materials were tested as interlayer materials such that their energy levels formed a cascade energy band structure and broadened the span of the solar spectrum absorbed by the photoactive layer.

2. Experimental methods

The devices were prepared on patterned indium tin oxide (ITO) glass substrates with 150 nm thick ITO and a 15 Ω /sq sheet resistance. After cleaning the ITO substrates, poly(3,4-ethylenedioxy thiophene)-doped poly(styrenesulfonate) (PEDOT:PSS) (Bytron P, from Bayer AG) was spin-coated onto the substrates at 4000 rpm for 60 s, followed by baking at 120 °C for 30 min. High-purity pentacene, phthalocyanine dyes such as Cu-phthalocyanine (CuPc)

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and metal-free phthalocyanine (H_2Pc), and C_{70} were provided by Nano C. Bathocuproine (BCP) was purchased from Aldrich. All organic layers were deposited on the ITO substrate by vacuum evaporation under a vacuum of 10^{-6} Torr. A pentacene layer (50 nm) was deposited at 0.2 \AA/s followed by a 10 nm thick phthalocyanine at 1.0 \AA/s and a 25 nm thick C_{70} at 0.5 \AA/s . An 8 nm BCP layer was deposited at 1.0 \AA/s as a hole blocking layer, and finally an 100 nm thick Ag layer was deposited as a cathode. The devices were characterized by current density–voltage (J – V) characteristics measured under AM 1.5 solar illumination at 100 mW/cm^2 generated by an Oriol 1 kW solar simulator using a programmable Keithley mode 4200 power source.

3. Results and discussion

The photovoltaic device structure and the corresponding energy band diagrams are shown in Figure 1. The structures of the photoactive layer are the bilayer structure, pentacene (50 nm)\(C_{70} (25 nm), and the three-layered structure, either pentacene (50 nm)\(H_2Pc or CuPc (10 nm)\(C_{70} (25 nm). Although the phthalocyanine layer was inserted between the pentacene and C_{70} layers in the three-layered structure, band offsets at all interfaces in the three-layered structure were comparable to the exciton binding energy (0.4–1.4 eV) [20] and efficient exciton-dissociation occurred due to the large band offset between the energy levels of the pentacene and C_{70} . The HOMO and LUMO levels of CuPc and H_2Pc were positioned, respectively, between the energy levels of the pentacene and C_{70} to form a cascade energy structure without energy barriers to charge transport toward the electrodes. The LUMO level of H_2Pc was slightly deeper than that of CuPc such that the LUMO offset at the pentacene\(H_2Pc interface matched the exciton binding energy of pentacene (0.1–0.6 eV) [21,22]. This factor was expected when taking into account the much lower electron affinity (EA) of Cu (1.22 eV) compared to the EA of H_2Pc (5.35 eV) [23]. The addition of a Cu atom to the H_2Pc molecule introduced additional charges and decreased the overall EA of the resulting molecule (CuPc) [23]. The energy levels of HOMO and LUMO levels in the organic materials were consistent with the values cited in the literature [23,24].

The current density–voltage (J – V) characteristics of the devices under illumination are shown in Figure 2 and Table 1. The open-circuit voltages (V_{OC}) of the devices with three photon-harvesting layers were higher than those of the device with two photon-harvesting layers. The V_{OC} depends on the energy difference between the LUMO level of the n -type material and the HOMO level of the p -type material [25]. The HOMO levels of H_2Pc and CuPc were similar and deeper than the level of pentacene, so that the V_{OC} values of the devices prepared with H_2Pc and with CuPc were not significantly different (0.40 V for H_2Pc , 0.39 V for CuPc) and higher than

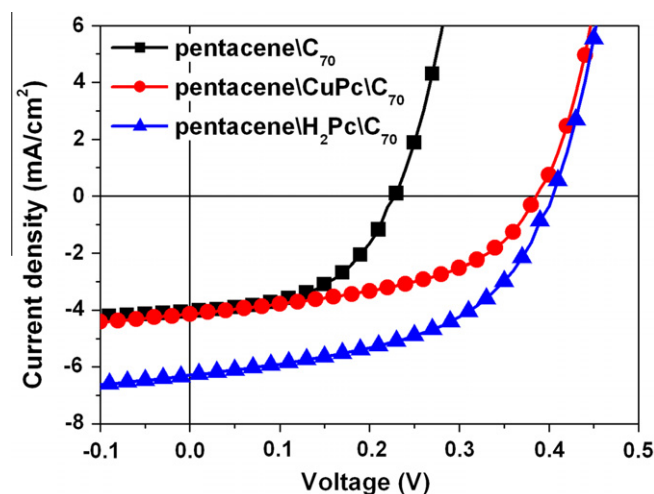


Figure 2. Current density–voltage characteristics under illumination for the three device structures: pentacene\(C_{70} , pentacene\CuPc\(C_{70} , and pentacene\(H_2Pc \(C_{70} .

Table 1

Summary of the device parameters associated with the three device structures tested here: pentacene\(C_{70} , pentacene\CuPc\(C_{70} , and pentacene\(H_2Pc \(C_{70} .

Device structure	J_{SC} (mA/cm^2)	V_{OC} (V)	FF	η (%)
Pentacene (50 nm)\(C_{70} (25 nm)	4.05	0.23	0.50	0.47
Pentacene (50 nm)\CuPc (10 nm)\(C_{70} (25 nm)	4.13	0.39	0.48	0.77
Pentacene (50 nm)\(H_2Pc (10 nm)\(C_{70} (25 nm)	6.29	0.40	0.51	1.28

the values of a device without phthalocyanine (0.23 V). The J_{SC} value of a device without phthalocyanine and a device with CuPc were nearly identical, and the J_{SC} of the device with H_2Pc exceeded the values of the other devices. The additional exciton-generation in the phthalocyanine layer contributed to the photocurrent of each device with three photon-harvesting layers; however, the negligible enhancement in J_{SC} value for the device prepared with CuPc may suggest that the additional CuPc layer limited charge generation from the pentacene layer. The higher value of J_{SC} in the device with H_2Pc suggested that charge separation in the device with H_2Pc was efficient, and the presence of an additional H_2Pc layer slightly hindered charge generation in the pentacene layer. The small changes in the FF demonstrated that the inserted phthalocyanine layer did not significantly affect the series resistances of the devices. As a result, the power conversion efficiencies (η_p) of devices with three photon-harvesting layers were higher than that of a device with two photon-harvesting layers.

Figure 3 shows the absorption spectra and external quantum efficiency (EQE) for each device structure and explains the increased J_{SC} in terms of the presence of an additional photon-harvesting layer with an appropriate energy level. The additional phthalocyanine layer absorbed longer wavelengths (700–800 nm) of the solar spectrum that pentacene and C_{70} could not absorb, as shown in Fig 3a and b. As a result, the layer increased the light absorption efficiency of the device. This wider light absorption range additionally increased the EQE over the range 700–800 nm, corresponding to the phthalocyanine absorption spectrum, as shown in Figure 3c. The additional excitons generated in the phthalocyanine layer contributed to the photocurrent. Excitons formed in the phthalocyanine were dissociated both at the pentacene\phthalocyanine interface and at the phthalocyanine\(C_{70} interface; however, the LUMO level difference at the phthalocyanine\(C_{70} interface (1.1 eV for CuPc and 0.95 eV for H_2Pc) was much higher than the HOMO level difference at the pentacene\phthalocyanine

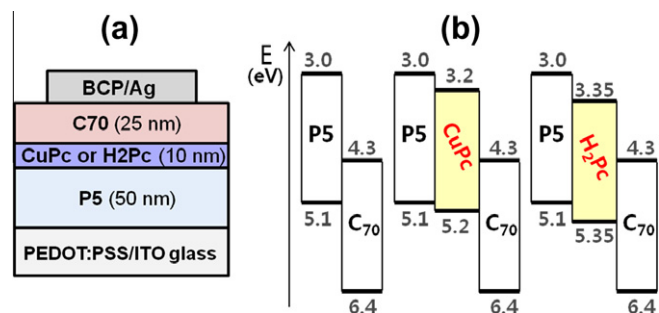


Figure 1. (a) Schematic diagram of the device structure. (b) Energy band diagrams of the devices with pentacene (P5) (50 nm)\(C_{70} (25 nm), P5 (50 nm)\CuPc (10 nm)\(C_{70} (25 nm), and P5 (50 nm)\(H_2Pc (10 nm)\(C_{70} (25 nm).

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