

Improved photovoltaic properties of pentacene/*N,N'*-Dioctyl-3,4,9,10-perylenedicarboximide-based organic heterojunctions with thermal annealing

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ABSTRACT

We have studied the effect of thermal annealing on the photovoltaic properties of an organic solar cell based on an efficient donor/acceptor combination of pentacene / *N,N'*-Dioctyl-3,4,9,10-perylenedicarboximide (PTCDI-C₈) discrete heterojunctions. As deposited samples showed a maximum power conversion efficiency about 1.12% under standard A.M 1.5 illumination, whereas 1.60% power conversion efficiency has been achieved after annealing the samples at 100 °C for 5 min. External quantum efficiency (EQE) is also found to be increased from ~60% to ~70% after heat treatment. Photocurrent action spectra and the absorption profile of this donor/acceptor combination confirm the efficient light harvesting capability of the device throughout the visible region of the solar spectrum. Morphology and crystallinity of the active layers improved significantly after annealing the samples, resulting in an almost two times enhancement in the short-circuit current density and improved overall cell performances.

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1. Introduction

In the last few decades, continuously increasing demand of alternative renewable energy sources has stimulated new scientific researches in the field of photovoltaic devices. Organic solar cells (OSCs) have attracted much attention as a promising choice for unconventional energy source because of their low-cost potential, light weight, mechanical flexibility and ease of processing [1–5]. In the last ten years, the power conversion efficiency of organic photovoltaic (OPVs) cells has significantly improved due to the use of new materials and precise device architectures. To date, both bilayer and bulk-heterojunction OPVs have achieved power conversion efficiencies up to ~5–6% [6–10] and efficiency about ~6.5% has been successfully realized in tandem OPV devices [11–12]. In addition to these, one of the major important factors for the successful realization of OPVs in real life application is the long operational lifetimes [13–17]. However the power conversion efficiency (η) and the stability of OSCs are still lower than inorganic or dye-sensitized solar cells. The performance of these solar cells has been limited due to the relatively short-exciton diffusion lengths, low charge carrier mobility and poor morphology of the active layers. For further improvement of OPVs, the short-circuit current density (J_{SH}) of the devices has to be

optimized accurately. The performance of OPVs has been found to be strongly depending on the microstructures of the active layers. Several researchers have shown that the device performance can be significantly enhanced by annealing the cells at different temperatures at different conditions [8], [18–21]. The improvement was attributed to the change in active layer crystallinity and morphology. Though thermal treatments have been proved to be very effective for bulk-heterojunction OPVs, surprisingly, similar effects have been rarely investigated for discrete heterojunction OPVs [22].

In this paper we have investigated the effect of annealing temperatures on the photovoltaic properties of pentacene/*N,N'*-Dioctyl-3,4,9,10-perylenedicarboximide (PTCDI-C₈)-based organic heterojunctions. We select pentacene and PTCDI-C₈ as donor and acceptor materials because of their high hole ($> 1 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for pentacene) and electron ($0.6 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for PTCDI-C₈) mobility [23–26] and better spectral coverage throughout the visible spectrum [27–28]. The device showed improved performance after annealing at 100 °C for 5 min.

2. Experimental procedures

2.1. Materials

In our study, we have used pentacene (99.9%) as an electron donor material and *N,N'*-Dioctyl-3,4,9,10-perylenedicarboximide

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(PTCDI-C₈) (98%) as an electron acceptor material. We have also used 2.8 wt% water solution of Poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) as hole transporting layer (HTL) and Tris-(8-hydroxyquinolato) aluminum (AlQ₃) (99.995%) as electron transporting layer (ETL). The chemical structures of pentacene and PTCDI-C₈ are shown in Fig. 1(a). All these organic materials were purchased from Sigma-Aldrich.

2.2. Film and device fabrications

The photovoltaic devices were fabricated on patterned indium tin oxide (ITO)-coated glass substrates. The sheet resistance of the ITO anode was 10 Ω/□. Prior to organic layer deposition, ITO substrates were cleaned thoroughly in acetone, isopropyl alcohol and de-ionized water in sequence using an ultrasonic cleaner followed by drying with nitrogen gas. Cleaned ITO anodes were treated with microwave oxygen plasma for 30 s. A 40-nm-thick PEDOT:PSS layer was spin coated on ITO anodes from a 2.8 wt% water solution. Spin-coated PEDOT:PSS substrates were annealed at around 110 °C for 1 h to remove the solvent completely. A 50-nm-thick pentacene layer was thermally evaporated with a deposition rate of ~0.5 Å/sec followed by the deposition of a 40-nm-thick PTCDI-C₈ layer. Annealing was performed by placing the samples inside the annealing chamber at a base pressure of 5×10^{-6} mbar for 5 min. The samples were then allowed to cool to room temperature before any further depositions. A 10-nm-thick electron transporting layer (ETL) of AlQ₃ was then deposited on the top of the PTCDI-C₈ layer [29–30]. Finally, an 80-nm-thick aluminum top electrode strip was deposited by thermal evaporation through a shadow mask to define a square-shaped active area of 4 mm². Base pressure during the thermal evaporation was 5×10^{-6} mbar.

2.3. Measurements

The thickness of different layers was monitored by quartz crystal monitor and was verified by stylus profilometer (Veeco

Dektak3). Atomic force microscopy (AFM) (Veeco Nanoscope-IV) in tapping mode was used to study the surface morphology of different layers of the device. The crystallinity of the pentacene/PTCDI-C₈ bilayer was studied using a Philips X-Pert PRO MRD X-ray diffractometer with CuKα radiation ($\lambda=0.15418$ nm). The absorption spectra of the pentacene/PTCDI-C₈ heterojunctions were investigated in the wavelength range 300–1100 nm using a UV-vis-NIR spectrophotometer (Perkin-Elmer Lambda 45). Current density–voltage (J–V) characteristics of the devices were measured using a Keithley 4200-SCS measurement unit in dark and under AM 1.5 illumination (Newport 67005 solar simulator) with a power density of 100 mW/cm². The photocurrent action spectrum was measured under short circuit condition at a chopping frequency of 180 Hz with a lock-in amplifier (Standford research system-SR830 DSP) during illumination with the monochromatic light from a broadband source. Photoluminescence lifetime decay measurements were done using Edinburgh Instruments (Life Spec-II, EPL 405) measurement unit with a 404.4 nm excitation pulse diode laser. All the device characterizations have been carried out under ambient condition.

3. Results and discussion

Materials used in photovoltaic devices should ideally have a strong absorption capability of solar energy, high-charge carrier mobility and large-exciton diffusion length. Their relatively short-exciton diffusion lengths and poor charge transport properties have limited the performance of OPVs. Another major limitation of organic PV energy conversion is the limited overlap between the active layer absorption with the solar spectrum, which limits the light harvesting efficiency of the device. Pentacene and PTCDI-C₈ can be an attractive choice as an efficient donor/acceptor combination for excellent charge transport properties and greater matching with the solar spectrum, as they showed significant absorption in the complementary part of the visible region. Fig. 1(a) shows the chemical structure of pentacene and PTCDI-C₈. Fig. 1(b) represents the absorption spectra of pentacene and PTCDI-C₈ films and pentacene/PTCDI-C₈-based heterojunctions. PTCDI-C₈ shows strong absorption in the wavelength range 400–600 nm with peaks at 488, 523 and 567 nm whereas pentacene shows significant absorption in the range of 500–700 nm with peaks at 583 and 667 nm and visible kinks at 501, 543 and 629 nm. Altogether, the pentacene/PTCDI-C₈ heterojunction shows excellent light harvesting ability throughout the visible solar spectrum.

Fig. 2(a) shows the J–V characteristics of ITO/PEDOT:PSS/pentacene (50 nm)/PTCDI-C₈ (40 nm)/AlQ₃ (10 nm)/Al devices under dark for different annealing temperatures. The device exhibits conventional rectifying diode characteristics as expected from any heterojunction devices sandwiched between electrodes having different work functions. The overall shape of the J–V curve under forward bias does not change considerably with annealing temperature, suggesting that there is very little modification or band bending at the interfaces. On the other hand, reverse bias current decreases with increasing annealing temperatures which leads to a significant improvement of the device rectification ration from $\sim 0.8 \times 10^1$ to $\sim 2.0 \times 10^2$ (at ± 2 V). This is mainly due to the elimination of microscopic shorts within the active layers and increase in shunt resistance on heating the devices at high temperatures.

The J–V characteristics under AM 1.5 (100 mW/cm²) simulated solar irradiation conditions of the same devices with different annealing temperatures are displayed in Fig. 2(b). The untreated samples exhibit a short-circuit current density (J_{sc}) of 3.40 mA/cm², an open-circuit voltage (V_{oc}) of 0.65 volt, and a fill

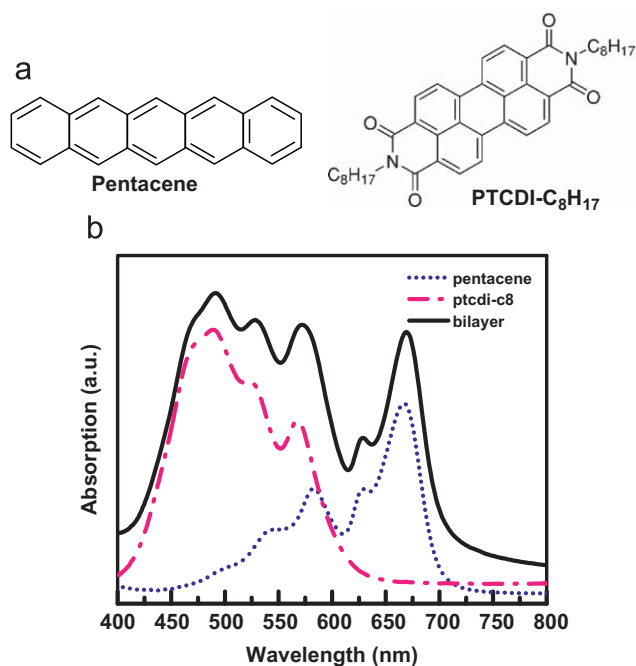


Fig. 1. (a) The chemical structure of pentacene and PTCDI-C₈. (b) Absorption spectrum of a 50 nm thick pentacene film, 40 nm thick PTCDI-C₈ film and the pentacene (50 nm)/PTCDI-C₈ (40 nm) heterojunctions deposited on glass substrates.

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