



Boosting electron extraction of inverted polymer solar cells using solution-processed nanocrystals as cathode interlayer



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ARTICLE INFO

Article history:

Received 2 May 2017

Received in revised form

8 November 2017

Accepted 12 November 2017

Available online 15 November 2017

Keywords:

Nanocrystals

Interlayer

Electron transport

Power conversion efficiency

Light absorption

ABSTRACT

In this paper, a powerful photoconductive interfacial layer was achieved by employing Cd₂SSe/ZnS quantum dots (CSSQDs) to modify the electron-transport layers (ETLs) in polymer solar cells (PSCs). With the optimal concentration of CSSQDs and superior film process condition, the power conversion efficiency (PCE) was increased from 4.12% to 6.49% for P3HT:ICBA based device, leading to a tremendous 1.6-fold performance enhancement. Moreover, the interlayer of CSSQDs could effectively improve the light absorption, the electron transport and extraction, as well as the better interfacial contact between the ETLs and active layers, accordingly resulting in a higher short-circuit current density, fill factor, and a lower series resistance, respectively. Hence, the introduction of CSSQDs interfacial layer could obtain a remarkably increased performance for the inverted PSCs.

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1. Introduction

Recently, bulk heterojunction (BHJ) polymer solar cells (PSCs) have been one of the most remarkable progresses in energy fields [1–5]. Among the rest, the inverted polymer solar cells (i-PSCs) have attracted widespread attentions because of the potential advantages in low cost of fabrication [6], portable carry of flexible material [7], large area of manufacture with printing technology [8], and superior photochemical stability of construction [9]. However, it is undeniable that the lower power conversion efficiency (PCE) of PSCs has a serious disadvantage for applicable promotion in comparison with prime inorganic solar cells, which mainly originates from the existent deficiency of the open-circuit voltage (V_{oc}) associating with band gap (E_g) and the narrow light absorption spectrum of the donor/acceptor (D/A) materials [10–12]. The novel materials play a vital role on the performance of photovoltaic devices, D/A polymer active layers commendably absorb the photon energy of illumination and realize the exciton generation and charge carrier transport [13–18], which are the

crucial factor for the short-circuit current (J_{sc}). Hence, some photosensitizers have been doped into active layers to achieve further improvement toward increasing light path and wavelength range in the visible region, leading to a significant enhancement of J_{sc} [19–22]. Meanwhile, interfacial engineering also fulfills the accomplishment, the interfaces between electrodes and active layers strengthen the charge transport properties between donor and acceptor, triumphantly switching over to more electron-hole pairs and more balancing charge carrier transfer and aggregation [23,24]. Moreover, the interface morphology and feature also need to be optimized to guarantee the increase of charge transport and fill factor (FF). Generally, the interfacial materials are rigidly selected to modify the transport layer, such as superior solubility, convenient processed procedure, and high light absorption [25]. Undoubtedly, the quantum dots (QDs) materials as excellent candidates could achieve prospective performance for i-PSCs, since they have adjustable and prolonged absorption spectra in the visible region, serving as exciton dissociation center and providing permeability channels between the active layers and electrodes, which could effectively promote the exciton diffusion and charge mobility [26,27]. In last few years, the core/shell structured QDs, whose cores are covered by the external shell materials, such as CdS/ZnS [28], are drawing more and more attention, which could

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dramatically reduce surface defect density and enhance stability for QDs construction [29,30].

In this contribution, the i-PSCs were designed and fabricated by introducing the Cd₂SSe/ZnS quantum dots (CSSQDs) to modify the polymer polyethylenimine (PEI) buffer layer, which based on poly(3-hexylthiophene) (P3HT) donor blending with indene-C60 bisadduct (ICBA) acceptor as active layers [31–33]. Consequently, the performance of the inverted cells was achieved an apparent improvement, and PCE of i-PSCs was increased from 4.12% up to 6.49% for the control device and optimal device, accompanying with 8.63–11.84 mA/cm² for *J*_{sc} and 56.79%–65.23% for FF. The results prove that CSSQDs could be one of the most potential candidates to modify the transport layer because of the prominent electrical and optical properties in light absorption, exciton generation and dissociation, charge transportation, and carrier mobility for the future photovoltaic application [34–36].

2. Experimental section

All reagents unless otherwise stated were purchased from 1-Material Inc. and Xi'an p-OLED Corp. and were applied as received without further purification. Firstly, the sculptured indium tin oxide (ITO)-glass substrate with superior light transmittance and matched energy level with electron-transport layers (ETLs) were ordinarily cleaned by acetone, alcohol, deionized water, and then desiccated by a nitrogen stream as cathode. Secondly, the PEI solution was ultrasonic treated about 30 min and uniformly spin-coated at 4000 rotations per minute (rpm) for 60 s and annealed in glovebox at 100 °C for 10 min. Thirdly, the prepared CSSQDs was dissolved in 1,2-dichlorobenzene with the concentrations of 0.5, 1.0, 1.5 mg/mL and then spin-coated onto PEI layer under 2500 rpm for 20 s, thus, the fabricated devices were named as Device B to D, respectively. Similarly, the control device without CSSQDs interlayer was named as Device A. The active layer blend with 1:1 wt ratio of P3HT:ICBA were spin-coated onto the CSSQDs interlayer and transferred into the glovebox for annealing at 150 °C for 25 min. Finally, a 10 nm of MoO₃ and a 100 nm of Ag were formed onto the active layers orderly by thermal evaporation in a vacuum chamber. The complete device structure of glass/ITO/PEI/CSSQDs/P3HT:ICBA/MoO₃/Ag and energy levels of the i-PSCs were presented in Fig. 1a and b as well as some implicated chemical structures of materials used in this study. MoO₃ has been proved as an efficient hole-transport layers in organic solar cells [37–39], which can facilitate the charge collection and extraction via decreasing interfacial carrier recombination, yielding the good hole transport and collection for the Ag electrode [40,41].

The CSSQDs were synthesized according to the previously reported non-injection processes [42,43]. Firstly, 40% Se nanocrystal,

1 mmol of cadmium acetate dihydrate, 15 mL of 1-Octadecene (ODE) and 2.4 mL of oleic acid were heated at 120 °C for 30 min in vacuum. The obtained liquid solution was cooled under N₂ flow to 30 °C and added a dry mixture of 0.2 mmol of Se and 0.3 mmol of S degassed in a three-neck round-bottom flask. The reaction mixtures were increased to 240 °C under N₂ environment with a rate of 10 °C per min. When the temperature rose to 240 °C, the reaction would begin. Nanocrystals were permitted to grow for 3 min after the reaction was quenched by injecting 8 mL of ambient temperature ODE. The product was cooled to room temperature by removing the heat source. Furthermore, the ZnS cap was grown on Cd₂SSe cores at 200 °C by dropwise adding an appropriate amount of a solution of diethylzinc and hexamethyldisilthiane in trioctylphosphine [44]. Also, the detailed synthetic process of CSSQDs used in our study is shown in Fig. 2a. The morphology of the CSSQDs was characterized using transmission electron microscopy (TEM) and the image of the product is shown in Fig. 2b. It can be seen that the diameter of CSSQDs is about 2–3 nm.

3. Results and discussion

Because the CSSQDs solution is spin-coated onto the PEI layer, we explored the optimal revolved condition in advance, the consistent density-voltage (*J*-*V*) characteristics were exhibited in Fig. 3a and detailed parameters are listed in Table 1, which were measured under AM1.5G illumination by Keithley 2601 source meter in normal environment [45,46]. The 1.5 mg/mL CSSQDs solution was prepared under 1500 rpm, 2500 rpm, and 3500 rpm for 20 s to produce homogeneous films in the identical external environment, proving that the 2500 rpm is the optimal parameter. Thus, the *J*-*V* characteristics of i-PSCs fabricated without and with the CSSQDs layer under the optimal spin-coating condition of 2500 rpm were shown in Fig. 3b, and all the performance values were summarized in Table 1. The experimental results between the different devices modified with CSSQDs could intuitively be observed from the Fig. 3b. It is worth noting that the control device (Device A) owns a common performance of the short-circuit current density (*J*_{sc}) of 8.63 mA/cm², an open-circuit voltage (*V*_{oc}) of 0.84 V, a fill factor (FF) of 56.79%, and a PCE of 4.12%, which is corresponding with the relative previous reported results [47,48]. The film thickness fabricated with the 1.5 mg/mL CSSQDs solution is about 11 nm, which would increase the probability of surface accumulation, additional trap states, charge recombination, and break the balance of the interface each other, leading to an unimpressive performance decrease of the device. When the concentration of CSSQDs is 0.5 mg/mL, the acquired film (5 nm) is not enough thick to modify the PEI layer to maximize the performance of device. Thus, the film fabricated using 1.0 mg/mL CSSQDs

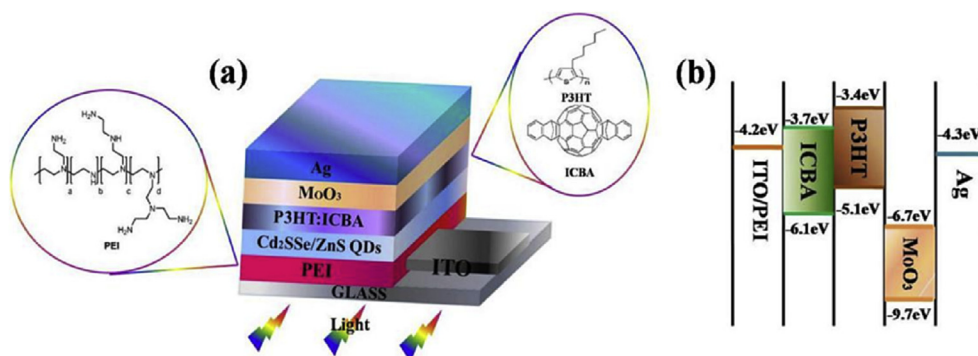


Fig. 1. (a) The device configuration and the chemical structure of used organic materials, and (b) the scheme of energy levels of the i-PSCs.

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