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Enhanced electron extraction capability of polymer solar cells via modifying the cathode buffer layer with inorganic quantum dots

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Enhanced performance of polymer solar cells (PSCs) based on the blend of poly[N-9’-heptadecanyl-2,7-carbazole-alt-5,5-(4’,7’-di-2-thienyl-2’-1’,3’-benzothiadiazole)] (PCDTBT):[6,6]-phenyl-C70-butyric acid methyl ester (PC71BM) is demonstrated by titanium dioxide (TiO2) interfacial modification via CuInS2/ZnS quantum dots (CZdots). Devices with a TiO2/CZdots composite buffer layer exhibit both a high short-circuit current density (Jsc) and fill factor (FF), leading to a power conversion efficiency (PCE) up to 7.01%. The charge transport recombination mechanisms are investigated by an impedance behavior model, which indicates that TiO2 interfacial modification results in not only increasing the electron extraction but also reducing impedance. This study provides an important and beneficial approach to develop high efficiency PSCs.

1. Introduction

Polymer bulk heterojunction (BHJ) solar cells based on composites of electron-donating polymers and electron-accepting fullerene derivatives exhibit potential advantages in making printable, portable, flexible, and large-area devices with relatively low-cost fabrication and are anticipated to be promising candidates to utilize solar energy.1–7 Over the last decade, many efforts have been devoted to further improving the performance of polymer solar cells (PSCs), such as designing new device structures and new polymers with enhanced properties and optimizing device morphology.8–14 However, the overall efficiency of BHJ-PSCs is still limited by many factors. The major drawbacks of BHJ-PSCs are often attributable to the narrow light absorption band of the polymers, low charge carrier mobility and limited exciton migration.15–20 Especially, a poor electron-extracting capability continues to be a big bottleneck for realizing high efficiency, and many approaches have been developed to improve charge carrier transfer for PSCs.

Recently, inverted structure PSCs with TiO2 or ZnO as a cathode buffer layer and high work function MoO3 as the anode buffer layer have attracted great attention because the inverted configuration provides better stability and design flexibility.21–23 However, the rebellious nanomorphology of the inorganic layer makes it difficult to attain an optimized interface with the polymer layer to enhance interconnected pathways for charge transport, as well as decrease charge recombination. For high performance inverted PSCs, the interfaces between electrodes and active layers should be an ohmic contact to maximize the open-circuit voltage (Voc) and minimize contact resistance. Furthermore, balanced charge transport and collection are basic requirements for highly efficient optoelectronic devices. Hence, various ways have been employed to modify the interfacial layer between the active layer and electrode to enhance charge transport and reduce the interfacial contact resistance.24,25 However, the choices of these ways used in polymeric materials are limited, as these require good compatibility in solubility, simple fabrication processes, and efficient charge transfer. An available strategy is to enhance the device efficiency by introducing soluble materials into the PSCs. Aqueous soluble inorganic quantum dot materials could also be promising candidates as efficient interface modification materials for highly efficient inverted PSCs,26 because they have superior charge carrier mobility and could act as exciton dissociation centers, which is beneficial for the future development of all-solution processed PSCs.27–29

In this contribution, we investigated the role of the CuInS2/ZnS quantum dots (CZdots) in interfacial modification of TiO2 for BHJ-PSCs. Impedance spectroscopy (IS) was examined in the dark with different bias voltages (including open-circuit conditions, Voc) to investigate the kinetics and energetic processes.

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governing the device performance. It is also an available tool to observe bulk and interfacial electrical properties that cannot be directly observed, such as the built-in voltage, doping concentration, diffusion time, and carrier mobility. By modelling the impedance behavior to investigate the charge transport recombination mechanisms, it was found that there is efficient charge transfer between TiO2 and the active layer, leading to improved electron mobility and energy level alignment for PSCs. The results demonstrate that CZdots can act as an efficient additive to improve the electron extraction capacity of BHJ-PSCs.

2. Experimental section

The CZdots nanocrystal (average diameter is about 4.5 nm and the surface ligand is oleic acid) was purchased from NNCrystal Co. Ltd. Poly[N-9’-hepta-decanyl-2,7-carbazole-alt-5,5-(4’,7’-di-2-thienyl-2’,1’,3’-benzothiadiazole)] (PCDTBT) and [6,6]-phenyl-C70-butryric acid methyl ester (PC71BM) were purchased from 1-Material Inc. and Lumtek Corp. without any purification, respectively. To fabricate inverted PSCs, commercial ITO-coated glass substrates were manually washed in acetone, isopropanol, and deionized water respectively. And then, the substrates were blow-dried with nitrogen and dealt with UV ozone for 20 min. The electron transporting layer (TiO2) was prepared by a sol–gel method, spin-coated at 3000 rotations per minute (rpm) for 20 s and annealed on a muffle furnace. The CZdots material was dissolved in chlorobenzene with a concentration of 2 mg mL⁻¹, and the solution was spin-coated on the surface of the TiO2 films. The devices modified with CZdots made at 3000, 2000, and 1000 rpm were named as Device B, Device C, and Device D, and the control device without CZdots was Device A. The corresponding film thicknesses of the CZdots layers made at 3000, 2000, and 1000 rpm are 4 nm, 7 nm, and 10 nm respectively, which were measured by an Ellipsometer. The active layer blend of PCDTBT:PC71BM was spin-coated on the CZdots layers and then annealed on a hot plate in a glove box at 70 °C for 20 min, and finally, the photovoltaic devices were finished by thermal evaporation of a thin layer of MoO3 (4 nm) followed by a Ag electrode (100 nm) with a furnace pressure lower than 10⁻⁵ Torr, and the active layer area of the device was defined by a shadow mask of 3.2 mm × 2 mm, so the device area of the PSCs is 6.4 mm². Except for the heat treatment of the active layer films in a glove box, all processes of device fabrication were done in air. All the data on the device performance presented in the article are typical average values of 32 devices from 8 repeated experiments.

3. Results and discussion

The effect of TiO2/CZdots as an electron transporting layer was investigated with the device structure of ITO/TiO2/CZdots/PCDTBT:PC71BM/MoO3/Ag, and the device structure and energy levels are shown in Fig. 1a and b respectively. Ultraviolet photoelectron spectroscopy (UPS) and a Kelvin probe system (KP) were combined to study the properties of the CZdots. It is worth noting that the TiO2/CZdots composite cathode buffer layer has a decreased work function compared to that of the pristine TiO2 film (Fig. 1c). As shown in Fig. 2a, UPS was used to measure the surface chemical composition and energy levels of the nanocrystals. The ultraviolet excitation energy (Ue) is 21.22 eV, where \( E_{\text{LUMO}} = U_e - (E_2 - E_1) \). \( E_1 \) is the side of the boundary on
the left and $E_2$ is on the right in Fig. 2a. The results indicate that the CZdots precursor solution spin-coating on TiO$_2$ substrates with proper temperature annealing, CZdots with a lowest unoccupied molecular orbital (LUMO) of 4.0 eV and a highest occupied molecular orbital (HOMO) of 7.6 eV have been formed, which is included in Fig. 1b. The work function of the CZdots was measured with a value of 4.2 eV, which matches well with the work function of TiO$_2$ and the LUMO of PC$_{71}$BM. In fact, the core/shell structure uniform distribution CZdots layer (Fig. 1d) plays a key role in electron transport from PC$_{71}$BM to TiO$_2$. As a result, this work function tuning by interfacial layers provides efficient charge collection centers and remarkably accelerates charge transfer rates from excited organic molecules to inorganic materials. Furthermore, the absorption and emission spectra of the CZdots are also located in the ultraviolet and visible region (Fig. 2b), and active layer absorption is presented in Fig. 2c, which shows that the emission spectrum of the CZdots is partially overlapping with the absorption area of the active layer. All of these increase the utilization of ultraviolet photons, which accounts for the enhancement of photon absorption for the PSCs (Fig. 2d). In order to prove the photovoltaic performance of the CZdots, Fig. 3a displays the representative current density versus voltage ($J$–$V$) characteristics, recording the performance of the PSCs prepared using a TiO$_2$ buffer layer with and without CZdots modification, and the corresponding photovoltaic average parameters of 32 devices are summarized in Table 1. It is noted that the device without the CZdots sensitized layer exhibited a relatively low efficiency, including a short-circuit current ($J_{sc}$) of 12.73 mA cm$^{-2}$, an open-circuit voltage ($V_{oc}$) of 0.86 V, a fill factor (FF) of 52.54%, and a calculated PCE of 5.72%. However, after the CZdots additive was employed to modify the TiO$_2$ interfacial layer, Device C shows a significant improvement with a $J_{sc}$ of 14.04 mA cm$^{-2}$, a $V_{oc}$ of 0.86, and a FF of 57.77%, leading to an increased PCE up to 7.01%, which is presented in Table 1. Observably, the surface treatment devices with an $x$ nm thick CZdots sensitization layer show a gradually improved performance, and the optimal thickness is $x = 7$ nm. It is also found that the device performance is highly sensitive to the thickness of the CZdots layer attached on the TiO$_2$ buffer layer, and the performance enhancement is ascribed to electron extraction capability improvement. Meanwhile, it can also be seen that the values of the $V_{oc}$ for the devices treated with CZdots have changed little compared to that of the control device, suggesting that CZdots have no effect on the built-in potential.

Fig. 2 (a) UPS image of the CZdots/TiO$_2$ film, (b) the absorption and emission spectra of the CZdots, (c) the absorption spectrum of the active layer, and (d) the absorption image of the completed devices.

Fig. 3 (a) $J$–$V$ characteristics of the solar cells without and with different thicknesses of CZdots, (b) IPCE spectra of all devices, (c) increasing rate of IPCE for the optimized devices, and (d) $J$–$V$ characteristics of all the photovoltaic devices in the dark.
To deeply investigate the mechanism of the $J_{sc}$ increase, the incident photon-to-electron conversion efficiency (IPCE) of the devices with and without the CZdots modifier was tested, and the obtained spectra are exhibited in Fig. 3b. As expected, the IPCE variation indicates a good agreement with the $J$–$V$ characteristics. Meanwhile, the IPCE of the devices modified with various thicknesses of CZdots shows a significant enhancement in a broad wavelength region of 375–750 nm compared to the control device, providing direct evidence for the gradually increased control device, providing direct evidence for the gradually increased control device, providing direct evidence for the gradually increased control device.

The IPCE difference data of the sensitized devices and the control device are shown in Fig. 3c, which is calculated by $R = (\text{IPCE}_{\text{CZdots device}} - \text{IPCE}_{\text{Control device}})/\text{IPCE}_{\text{Control device}}$. In order to further certify the $J_{sc}$ and FF increase, the dark $J$–$V$ characteristics of the PSCs are displayed in Fig. 3d. The modified devices with CZdots interfacial layers exhibit a smaller leakage current at a negative voltage and a low positive voltage, and the dark $J_{sc}$ increases sharply at a high positive voltage, resulting in a high rectifying ratio of the diodes. The improved dark $J_{sc}$ characteristics lead to a decrease of the series resistance and an increase of shunt resistance, contributing to the enhancement of the FF and PCE. The statistical histograms of the photovoltaic parameters for the control and the best optimized devices are exhibited in Fig. 4, and the $V_{oc}$, $J_{sc}$, FF, and PCE results are included in Fig. 4a–d.

<table>
<thead>
<tr>
<th>Device</th>
<th>$V_{oc}$ (V)</th>
<th>$J_{sc}$ (mA cm$^{-2}$)</th>
<th>FF (%)</th>
<th>PCE (%)</th>
<th>$R_s$ (Ω)</th>
<th>$R_{sh}$ (Ω)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0.86</td>
<td>12.73</td>
<td>52.50</td>
<td>5.96</td>
<td>224.14</td>
<td>4807.54</td>
</tr>
<tr>
<td>B</td>
<td>0.86</td>
<td>13.10</td>
<td>54.67</td>
<td>6.27</td>
<td>183.92</td>
<td>3764.81</td>
</tr>
<tr>
<td>C</td>
<td>0.87</td>
<td>14.04</td>
<td>57.77</td>
<td>7.19</td>
<td>133.38</td>
<td>24981.2</td>
</tr>
<tr>
<td>D</td>
<td>0.86</td>
<td>13.84</td>
<td>52.87</td>
<td>6.50</td>
<td>198.81</td>
<td>6757.36</td>
</tr>
</tbody>
</table>

To gain more insight into the role of the CZdots layer on the mechanism of charge recombination and transport process for the control and optimal devices, the dependence of the photocurrent density ($J_{ph}$) on the effective voltage ($V_{eff}$) on a double-logarithmic scale is indicated in Fig. 5, which was obtained without and with illumination of 100 mW cm$^{-2}$. Herein, $J_{ph} = J_{sc} - J_{0}$, and $J_{sc}$ and $J_{0}$ are the current densities under illumination and in the dark, respectively. Simultaneously, $V_{eff} = V_e - V$, $V$ is the applied voltage, and $V_e$ is the compensation voltage at $J_{ph} = 0$. $J_{ph}$ increases linearly with the effective voltage ($V_{eff}$) when $V_{eff}$ is less than 0.3 V, and the slope shows that the maximum exciton generation rates ($G_{max}$) of the sensitized solar cells are higher than that of the control device. It is worth noting that $J_{ph}$ increases faster with $V_{eff}$ in the optimized devices, which implies a faster rate of absorbed photons to electrons. Especially, interface modification increases the electron transport and collection, hence the response speed of the sensitized devices becomes faster, leading to an enhanced exciton dissociation probability ($P(E,T)$). $P(E,T)$ was calculated by normalized processing: $P(E,T) = J_{ph}/J_{sat}$, where $J_{sat}$ is the saturation photocurrent density. Fig. 5 shows that $J_{ph}$ tends to saturate at a sufficiently high value ($V_{eff} > 1$ V), and the higher $J_{ph}$ value usually implies a higher charge extraction efficiency, accounting for a relatively increased FF and $J_{sc}$ for the modified devices. $J_{sat}$ can be deduced from Fig. 5, which is only related to the absorbed incident photon flux and is independent of the bias and temperature. All of these characteristics are reflected in the advanced IPCE present in Fig. 3b, and the modified devices show higher IPCE values across the entire wavelength region from 300 to 800 nm compared to the reference device. The maximum IPCE values for the optimally modified and the control devices are 75% and 63% at 480 nm respectively, which match with the $J_{sc}$ trend well.

It is widely recognized that the nanomorphology of the films plays a crucial role in the charge separation and transport process for organic photovoltaic devices. The influence of surface modification on the TiO$_2$ buffer layer by CZdots was examined by atomic force microscopy (AFM). Fig. 6 shows the AFM topographic images of bare TiO$_2$ films as well as the TiO$_2$/CZdots films with different thicknesses. For Device C, it shows a very smooth and featureless structure with a low root-mean-square (RMS) value of 0.96 nm. However, the RMS values slightly increased to
5.54 nm (Device A), 1.04 nm (Device B), 1.37 nm (Device D), and 1.83 nm (CZdots film made under 500 rpm), respectively. It means that surface modification with CZdots changes the roughness of the TiO₂ cathode buffer layers. The minor variation of the film morphology would have a significant impact on charge transfer, and the smooth and featureless interface improves the electron transport and collection. The introduction of CZdots facilitates the electron injection and smaller charge recombination probabilities in the interface. Therefore, it is reasonable to speculate that the electron transport property will be energetically favored at the TiO₂/CZdots interface, which is confirmed by the J_sc enhancement of the modified devices (Fig. 6f).

Fig. 6 AFM morphology image of the TiO₂/CZdots films: (a) pristine TiO₂ film, (b) 4 nm CZdots layer, (c) 7 nm CZdots layer, (d) 10 nm CZdots layer, and (e) 15 nm CZdots layer. (f) J–V characteristics of the control and optimized cells under illumination and in the dark (inset).

Generally, efficient interfacial modification usually leads to lower resistance for photovoltaic devices, and the series resistance greatly affects the J–V characteristics of the PSCs. The J–V equation of the PSCs is usually modeled by means of a behavior as:

\[
j = \frac{1}{1 + \frac{j_{0}}{R_{p}}} \left\{ j_{0} \exp \left( \beta \frac{V_{app} - jR_{i}}{k_{B}T} \right) - 1 - \frac{J_{ph} - V_{app}}{R_{p}} \right\}
\]

where \(V_{app}\) is the applied voltage, and \(J_{ph}\) accounts for the photocurrent, which is reduced by the shunt resistance \(R_{p}\). Besides, \(k_{B}\) stands for the thermal energy, \(q\) is the elementary charge, \(j_{0}\) represents the dark current, and the parameter \(\beta\) accounts for the deviation from the diode ideal equation (inverse of the diode ideality factor). Therefore, an equivalent circuit diagram for the actual blending system of the organic solar cell in the actual test has been given, from which different circuit elements for the actual factors can be derived. As shown in Fig. 7a, \(C_{1}\) is the chemical capacitance (connected to carrier storage), \(R_{rec}\) is the recombination resistance (derivative of the carrier recombination flux) response, \(R_{i}\) is the transport-related resistance, \(C_{i}\) is related to geometrical and dielectric mechanisms, and \(R_{0}\) is the anode interface, buffer layer, and wire resistance for the PSCs. The series resistance \(R_{s}\) was introduced as \(R_{s} = R_{1} + R_{0}\). It can be seen from Table 1 that \(R_{s}\) significantly tends to decrease due to the incorporation of CZdots, resulting in an increased FF.

Fig. 8 presents the Nyquist plots of the impedance spectra for the control and modified devices at different thicknesses of CZdots at the open-circuit applied voltage. These semicircles can help us to understand the internal resistance of the different modified devices. From the impedance spectra, it clearly depicts that the semicircle’s diameter for the reference devices is much larger than that of the modified devices. It demonstrates the
recombination resistance of the CZdots modified devices is apparently decreased. Meanwhile, as shown in Table 1, the calculated series resistance from the $J-V$ curve is obviously reduced, which contributes to the increase of the $J_{sc}$ and FF. And a higher capacitance is obtained after interfacial modification with CZdots, resulting in a higher electron concentration.

These experimental data resemble the typical semicircle shape, which can be accurately modeled with a simple parallel RC circuit (Fig. 7c). The imaginary part and real part in Fig. 7b, mapped as a semicircle in the coordinate axes, are related to the resistance:

$$Z = R_1 + \frac{1}{\frac{1}{R_c} + j\omega C_\mu}$$

or

$$Z = R_1 + \frac{R_t}{1 \omega R_t C_d} - \frac{\omega R_t^2 C_\mu}{1 + (\omega R_t C_\mu)^2}$$

The data of the center coordinates of the circle formulate ($x = R_1 + R_t/2$, $y = 0$), and $R_1$, $R_t$ and $C_\mu$ can be given by the following equations:

$$OA = R_1$$

$$AC = R_t$$

$$C_\mu = \frac{1}{\omega \sigma_0 R}$$

$$i_0 = RTlnFR_t$$

The capacitance $C_\mu$ is attributed to the depletion region chemical capacitance due to connecting to the carrier storage.$^{54}$ This capacitance follows the Mott–Schottky expression:

$$C_\mu^{-2} = \frac{2(V_{bi} - V)}{A^2 \varepsilon_0 \varepsilon D N}$$

and

$$V_{bi} = \phi_{bi} + \frac{KT}{\varepsilon}$$

where $V_{bi}$ is the built-in potential, $N$ is the impurity density, $A$ is the area (6.4 mm$^2$ in this paper), $V$ is the applied voltage, and $\varepsilon_A$ and $\varepsilon_0$ are the dielectric constant and vacuum permittivity, respectively. In order to explore the role of the CZdots interfacial layer on the device performance improvement, the capacitance–voltage characteristics of the devices were measured and compared to the result obtained from the Mott–Schottky relation. The charge carrier density can be calculated by:

$$N = \frac{1}{e} A d \int_{V_{oc}}^{V} C(V) dV$$

where $e$ is elementary charge, $A$ is the device area, $d$ is the thickness of the active layer, and $C$ is the chemical capacitance. The chemical capacitance is simulated using the Schottky equivalent circuit model and following the Mott–Schottky expression. Fig. 9 shows the impedance spectra of all the devices at different voltage values. From the impedance spectra, we extracted $C_\mu$ and the carrier concentration (ND) from the linear region. For the optimized device (Fig. 9c), the average electron number is around $1.81 \times 10^{15}$, which is slightly higher than that obtained from the control device ($1.62 \times 10^{15}$ calculated from Fig. 9a). We also calculated the carrier concentrations for Device B (Fig. 9b) and

Fig. 7 (a) Equivalent circuit diagram for the actual blending system of the PSCs, (b) impedance spectra for the simplified circuit of the PSCs, and (c) simplified circuit diagram for the actual blending system of the PSCs.

Fig. 8 Impedance spectra of the PSCs with different thicknesses of CZdots under open-circuit conditions.

Fig. 9 Impedance spectra of (a) Device A, (b) Device B, (c) Device C, and (d) Device D at different applied voltage values, respectively.

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OA = $R_1$

AC = $R_t$

$C_\mu = \frac{1}{\omega \sigma_0 R}$

$i_0 = RTlnFR_t$

$V_{bi} = \phi_{bi} + \frac{KT}{\varepsilon}$

$N = \frac{1}{e} A d \int_{V_{oc}}^{V} C(V) dV$
Device D ([Fig. 9d]), which are also smaller than that of the optimized device. Except for the improved morphology, CZdots can be considered as an efficient electron transfer center, and CZdots could accelerate efficient exciton dissociation at the active layer interface. Likewise, it is reasonable to speculate that exciton dissociation would be energetically favored at the PCDTBT:PC71BM and TiO2 interface, which is confirmed by the increased electron number from the reference device to the optimal modified device.

The equivalent circuit diagram for the PSCs in the actual test shows that a capacitor connects the anode and the cathode. In fact, the AC voltage as a modulating signal is applied to the electrodes of the PSCs, which induces current crossing across the devices. This results in a capacitor-like device that stores and releases energy quickly. Thus, the electron mobility can be calculated from this process. In order to further probe the transport recombination mechanism for the optimized devices, admittance spectroscopy was measured to get the electron mobility and carrier transit time. Fig. 10 presents the capacitance–frequency characteristics of all the fabricated devices. In summary, a CZdots interface modified layer on TiO2 plays a significant role on the $J_{sc}$ and FF, leading to a great PCE enhancement. The feature of CZdots was studied using UPS and KP, and the effect of TiO2 interfacial modification via CZdots is demonstrated by the spectroscopy techniques and AFM. The application of CZdots interfacial modification changes the roughness of the electron transporting layer, resulting in a high electron mobility. Improved nanostructures of the TiO2 surface decrease interfacial charge traps between the active layer and TiO2, achieving a smaller charge recombination probability. Furthermore, CZdots can also serve as efficient charge transport channels to accelerate electron collection. This study demonstrates an effective approach to optimize the performance of PSCs using quantum dot materials.

4. Conclusions

In summary, a CZdots interface modified layer on TiO2 plays a significant role on the $J_{sc}$ and FF, leading to a great PCE enhancement. The feature of CZdots was studied using UPS and KP, and the effect of TiO2 interfacial modification via CZdots is demonstrated by the spectroscopy techniques and AFM. The application of CZdots interfacial modification changes the roughness of the electron transporting layer, resulting in a high electron mobility. Improved nanostructures of the TiO2 surface decrease interfacial charge traps between the active layer and TiO2, achieving a smaller charge recombination probability. Furthermore, CZdots can also serve as efficient charge transport channels to accelerate electron collection. This study demonstrates an effective approach to optimize the performance of PSCs using quantum dot materials.

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**Notes and references**


