



# CdTe quantum dots-sensitized solar cells featuring PCBM/P3HT as hole transport material and assistant sensitizer provide 3.40% efficiency

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## ABSTRACT

A heterojunction consisted of [6,6]-phenyl- $C_{61}$ -butyric acid methyl ester (PCBM) and poly(3-hexylthiophene) (P3HT) was employed as hole transporter and light absorber assistant, a microporous platinum/fullerenes (Pt/ $C_{60}$ ) counter electrode was prepared by using a facile thermal decomposition method, and a polydimethyldiallyl ammonium-cadmium telluride [(PDDA)-CdTe] quantum dots photoanode was prepared by using chemical bath deposition method. Based on above components, a CdTe quantum dot-sensitized solar cell (QDSSC) was fabricated. The QDSSC shows a light-to-electric energy conversion efficiency of 3.40% under a simulated solar light irradiation with an intensity of  $100 \text{ mW cm}^{-2}$ . The electrochemical and photovoltaic measurements indicate that microporous Pt/ $C_{60}$  film is better than Pt film as counter electrode material for the QDSSCs, and PCBM/P3HT is better than iodide/triiodide and sulfide/polysulfide as transferring medium for QDSSCs.

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## 1. Introduction

Since its prototype was reported in 1991 by Gratzel [1], dye-sensitized solar cell (DSSC) based on Ru complexes as sensitizer dye has attracted significant attention as a promising alternative to conventional Si solar cell. A light-to-electric energy conversion efficiency of 12% for the DSSC has been achieved [2,3].

Recently, inorganic semiconductor quantum dots (QDs, including CdS, CdSe, PbS, etc.) [4–6] have been extensively explored to replace metalorganic sensitizers for the preparation of QDs sensitized solar cells (QDSSCs) due to their high extinction coefficient, potential multiple exciton generation, spectral tunability by particle size, and high energy conversion efficiency for QDSSCs as 44% in theoretically [7–9]. However, the conversion efficiencies for QDSSCs reported were lower (typically below 3%), so far, which was due to their narrow absorption range and charge recombination at the QDs|electrolyte interface [10,11]. Many studies have shown that the iodide/triiodide ( $I^-/I_3^-$ ) electrolyte was chemically incompatible with QDs in QDSSCs [10]. The QDSSCs based on sulfide/polysulfide ( $S^{2-}/S_x$ ) electrolyte were reported to give a highest efficiency of 4.1% [11]. However, the device had poor fill factor ( $FF$ ) due to undesirable charge recombination between the electrolyte and accumulated electrons on the  $TiO_2$  surface incompletely covered by the QDs [12,13]. Thus, it is necessary to develop new

materials and replace the  $I^-/I_3^-$  and  $S^{2-}/S_x$  electrolyte to reduce charge recombination.

A blend heterojunction consisted of [6,6]-phenyl- $C_{61}$ -butyric acid methyl ester (PCBM) and poly(3-hexylthiophene) (P3HT) has been extensively investigated in polymer photoelectric devices [14,15], which can also be used as an efficient hole conductor and light absorber in the DSSCs [16]. The microporous platinum/fullerenes (Pt/ $C_{60}$ ) film can improve charge transfer and reduce charge recombination, which can be applied as counter electrode materials in DSSC.

In the present work, a  $TiO_2$ -polydimethyldiallyl ammonium (PDDA)-CdTe photoanode was prepared by using chemical bath deposition (CBD) method, a (Pt/ $C_{60}$ ) film is used as counter electrode, and a heterojunction PCBM/P3HT is applied as hole conductor and assistant light absorber. Based on above components, a CdTe QDSSC is fabricated. The device presents a broader spectral response in the region of 400–750 nm, improves charge transfer, reduces charge recombination and enhances light absorption, a high energy conversion efficiency of 3.40% is achieved for the CdTe QDSSCs under a simulated solar light irradiation with an intensity of  $100 \text{ mW cm}^{-2}$ .

## 2. Experimental

### 2.1. Materials

Titanium(IV)isopropoxide and 4-tert-butylpyridine (TBP) were purchased from Fluka and used as received. The anhydrous ethanol (ETOH), isopropanol, acetic acid (HAc), chloroplatinic acid

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( $\text{H}_2\text{PtCl}_6$ ), emulsification agent polyoxyethylene octyl phenol ether (OP, Triton X-100), polyethylene glycols with average molecular weights 20,000 (PEG-20000), tetrabutyltitanate [ $\text{Ti}(\text{OBU})_4$ ], thioglycolic acid (TGA), titanium tetrachloride ( $\text{TiCl}_4$ ), cadmium chloride ( $\text{CdCl}_2 \cdot 2.5\text{H}_2\text{O}$ ), tellurium powder (Te), potassium borohydride ( $\text{KBH}_4$ ), fullerenes ( $\text{C}_{60}$ ) and other reagents were all purchased from Shanghai Chemical Agent Ltd, China. Polydimethyl-diallyl ammonium chloride (PDDA) was purchased from Acros Co., USA. All reagents were used without further treatment before use.

A conductive glass plate (FTO glass, with fluorine doped tin oxide over-layer, sheet resistance of  $8 \Omega \text{ cm}^{-2}$ , purchased from Hartford Glass Co., USA) was used as a substrate for precipitation of the  $\text{TiO}_2$  porous film and was cut into  $1 \text{ cm} \times 2 \text{ cm}$  sheets.

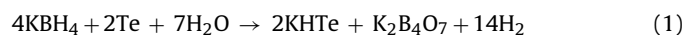
## 2.2. Preparation of microporous Pt/ $\text{C}_{60}$ counter electrode

Typical Pt/ $\text{C}_{60}$  counter electrode was prepared as following:  $\text{H}_2\text{PtCl}_6$  (1.0 wt.%) solution was prepared by dissolving  $\text{H}_2\text{PtCl}_6$  in a mixed solution of isopropanol and *n*-butanol (volume ratio of 1:1) containing 2–3 drops of OP emulsification agent. The  $\text{C}_{60}$  (0.3 wt.%) was dispersed in the  $\text{H}_2\text{PtCl}_6$  solution. A cleaned FTO substrate was heated for 10–15 min to reach a suitable temperature (200–250 °C) in atmosphere environment. Then the  $\text{H}_2\text{PtCl}_6$ : $\text{C}_{60}$  solution was rapidly dropped on the heated FTO substrate surface, which resulted in the rapid decomposition of  $\text{H}_2\text{PtCl}_6$  and the formation of Pt/ $\text{C}_{60}$  film. The film was sintered at 450 °C for 30 min to remove some organic compounds remained, thus a microporous Pt/ $\text{C}_{60}$  counter electrode was prepared.

For comparison, conventional Pt counter electrode was also prepared by dropping some  $\text{H}_2\text{PtCl}_6$  isopropanol solution on the FTO substrate and heated at 450 °C for 30 min under air ambient [17,18].

## 2.3. Synthesis of CdTe quantum dot

Briefly, preparation of CdTe QDs was as following [19,20]: 4 mM  $\text{KBH}_4$  and 2 mM Te powder were dissolved in appropriate amount of deionized water under high-purity nitrogen protection for 15 min. The mixture solution was stored at 4 °C for 8 h in refrigerator, when a chemical reaction takes place, as shown below, KHTe purple transparent solution was obtained.



A series of CdTe aqueous colloidal solutions were prepared by adding freshly KHTe solution to  $1.25 \times 10^{-3} \text{ N}$   $\text{CdCl}_2$  solutions at pH 10.2 in the presence of thioglycolic acid (TGA) as a stabilizing agent under  $\text{N}_2$  atmosphere. The molar ratio of  $\text{Cd}^{2+}$ /stabilizer/ $\text{HTe}^-$  was fixed at 1:2.4:0.5. The resultant mixture was then subjected to a reflux to control the nanocrystal growth of CdTe at 100 °C. CdTe

samples with different particle sizes were obtained by controlling different reflux times.

## 2.4. Fabrication of QDSSCs

The porous  $\text{TiO}_2$  photoanode was prepared as described previously [14,21]. A thin  $\text{TiO}_2$  blocking layer was deposited on the FTO substrate by immersing the FTO in 0.15 M  $\text{TiCl}_4$  isopropanol solution for 12 h, followed by sintering at 450 °C for 30 min in air. Subsequently, a  $\text{TiO}_2$  layer with a particle size of 10–20 nm and thickness of 10  $\mu\text{m}$  was coated on the blocking layer by using a doctor blade method, followed by sintering at 450 °C for 30 min in air. Thus a  $\text{TiO}_2$  anode was obtained. The  $\text{TiO}_2$ -PDDA electrode was prepared using chemical bath deposition (CBD) method. In brief,  $\text{TiO}_2$  electrode with an effective area of  $0.2 \text{ cm}^2$  was immersed in PDDA (1 wt.%) solution for 30 min, dried with an air gun, dipped for another 30 min into a fresh CdTe solution in a vial heated at 50 °C, and then dried with an air gun. The processes were repeated five times (shown in Fig. 1) to prepare brown  $\text{TiO}_2$ -PDDA-(CdTe) $_n$  electrode, where *n* is an integer from 1 to 5. The CBD method was also used to deposit the ZnS passivation layer. The electrode was dipped sequentially into a 0.5 M  $\text{Zn}(\text{NO}_3)_2$  ethanol solution and a 0.5 M  $\text{Na}_2\text{S}$  methanol solution. The optimal dipping time was found to be 1 min for both solutions.

PCBM:P3HT was spin-coated at 2000 rpm from its chlorobenzene solution of the PCBM and P3HT (1:2, w/w) with the polymer concentration of  $15 \text{ mg mL}^{-1}$  on  $\text{TiO}_2$ -PDDA-(CdTe) $_n$  film electrode and dried at 150 °C for 10 min, giving a PCBM/P3HT thickness of ca. 90 nm [14,22]. Finally, the Pt/ $\text{C}_{60}$  counter electrode was pressed on the PCBM/P3HT layer and a QDSSC was sealed by using a cyanoacrylate adhesive as a sealant to insulate from the air and water outside. The detailed fabrication procedure for the DSSC was described by us elsewhere [22–24].

## 2.5. Characterization and measurements

The micromorphology of the Pt/ $\text{C}_{60}$  counter electrode and the  $\text{TiO}_2$ /PDDA/CdTe photoanode were observed by using an S-4800 field emission scanning electron microscope. The ultraviolet-visible (UV-vis) absorption spectra of samples were measured using an Optizen (3100 UV) spectrophotometer. The electrochemical impedance spectroscopy (EIS) was carried out using a CHI660D electrochemical measurement system at a constant temperature of 20 °C with AC signal amplitude of 20 mV in the frequency range from 0.1 to  $10^5 \text{ Hz}$  at 0 V DC bias in the dark. The photoluminescence (PL) of the samples were measured using an F-4500 fluorescence spectrophotometer (Hitachi Corporation, Japan).

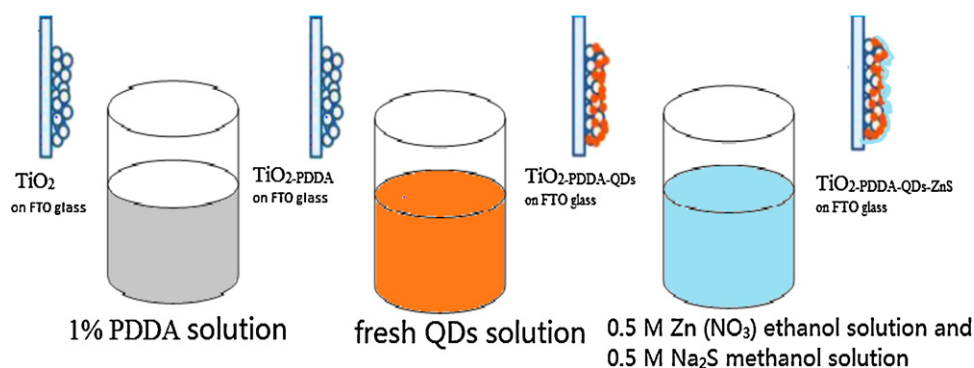


Fig. 1. The schematic of the  $\text{TiO}_2$ -PDDA-(CdTe) $_n$  electrode prepared by CBD method. (For interpretation of the references to color in the text, the reader is referred to the web version of this article.)

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