



# CdHgTe and CdTe quantum dot solar cells displaying an energy conversion efficiency exceeding 2%

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## ABSTRACT

We have fabricated quantum dot solar cells (QDSSCs) incorporating CdHgTe nanocrystals (NCs) and CdTe quantum dots (QDs), which we prepared separately from aqueous mixtures of NaHTe, Cd(NO<sub>3</sub>)<sub>2</sub>, and 3-mercaptopropionic acid in the presence and absence of HgCl<sub>2</sub>, respectively. After depositing the CdHgTe NCs and CdTe QDs onto TiO<sub>2</sub> electrodes coated with poly(dimethyldiallylammonium chloride), stabilized through electrostatic interactions, we obtained (CdHgTe)<sub>3</sub> and (CdHgTe)<sub>3</sub>-(CdTe)<sub>2</sub> QDSSCs having energy conversion efficiencies of 1.0% and 2.2%, respectively. The incident photon-to-current conversion efficiencies (IPCEs) of the (CdHgTe)<sub>3</sub> and (CdHgTe)<sub>3</sub>-(CdTe)<sub>2</sub> QDSSCs were 12.2% and 17.5%, respectively. The higher energy conversion efficiency of the (CdHgTe)<sub>3</sub>-(CdTe)<sub>2</sub> QDSSCs relative to that of the (CdTe)<sub>3</sub>-(CdHgTe)<sub>2</sub> QDSSCs resulted from the greater electron transfer efficiency in the former system. The combination of CdHgTe NCs and CdTe QDs and their deposition sequence are two important factors affecting the QDSSCs' efficiencies, which were controlled by their photocurrent densities and fill factors.

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## 1. Introduction

Quantum dot-sensitized solar cells (QDSSCs) have great potential to become efficient energy generators [1–3]. Although QDSSCs provide lower energy conversion efficiencies ( $\eta$ ) than those of single-crystal SiO<sub>2</sub>-based chips ( $\eta > 20\%$ ) and dye-sensitized solar cells ( $\eta < 10\%$ ) [4], they are easy to prepare cheaply. Relative to the dyes used in solar cells, QDs offer several advantages: photostability, greater molar extinction coefficients, multi-exciton generation capability, and size-dependent optical properties [1–3]. Much effort has been devoted to the fabrication of QDSSCs by harnessing small-bandgap semiconductors (e.g., CdSe, PbS, CdS, and InP) as sensitizers because they can transfer electrons to large-bandgap semiconductors (e.g., TiO<sub>2</sub>) when illuminated with sunlight [5–8]. The values of  $\eta$  of most reported QDSSCs are, however, less than 2.0%, mainly because of inefficient electron transfer, a narrow range of absorption wavelengths in the solar spectrum, and non-radiative decay [5–11].

The  $\eta$  in QDSSCs can be further improved by using mixtures of several different types of QDs to widen the absorption coverage of the solar spectrum [12–14]. A co-sensitized QDSSC consisting of two differently sized CdSe QDs, which were adsorbed onto TiO<sub>2</sub> nanotubes, provided a value of  $\eta$  of 1.20% [15]. QDSSCs fabricated from three different sizes of CdTe QDs on TiO<sub>2</sub> nanotubes exhibited increased degrees of light harvesting and higher-efficiency electron injection [16]. In these QDSSCs, smaller-sized

QDs are employed to maximize the transfer of electrons to the TiO<sub>2</sub> electrode, while the larger ones capture additional incident light. QDSSCs fabricated from multilayered CdS/CdSe/ZnS QDs on TiO<sub>2</sub> electrodes through chemical deposition have provided a noteworthy value of  $\eta$  of 4.2% [17].

Recently, we developed a thermal strategy for the fabrication of QDSSCs from CdTe QDs [18]. After thermally treating CdTe QDs adsorbed onto TiO<sub>2</sub> electrodes through three cycles, each for a different period of time, differently sized CdTe QDs were deposited onto the substrate coated with TiO<sub>2</sub> nanoparticles (NPs). The as-prepared QDSSCs provided superior values of the open-circuit photovoltage ( $V_{OC} = 850$  mV) and  $\eta$  (2.02%) at 100% sunlight (AM 1.5, 100 mW/cm<sup>2</sup>), although the photocurrent density ( $J_{SC}$ ) was lower than those of other reported QDSSCs. In this study, we used CdHgTe NCs and CdTe QDs, instead of differently sized CdTe QDs, to prepare QDSSCs. Here, we focused on determining the impact that the coating sequence of the two different nanomaterials on substrates treated with TiO<sub>2</sub> NPs had on the performance of the QDSSCs with respect to their values of  $\eta$ ,  $V_{OC}$ ,  $J_{SC}$ , and fill factor (FF). When optimized, the (CdHgTe)<sub>3</sub> and (CdHgTe)<sub>3</sub>-(CdTe)<sub>2</sub> QDSSCs provided values of  $\eta$  of 1.0% and 2.2%, respectively.

## 2. Experimental

### 2.1. Chemicals and instruments

Commercially available cadmium nitrate (99%), 1-ethyl-3-methylimidazolium thiocyanate (EMImSCN), ethylene glycol (99%),

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mercury chloride, hydrogen hexachloroplatinate(IV) (99%), methyl cellulose, 3-MPA (99%), poly(dimethyldiallylammonium chloride) ( $M_w$  400,000–500,000, 20 wt% aqueous solution), polyethylene glycol ( $M_w$  5000), poly(vinylpyrrolidone) (PVP,  $M_w$  55,000), P-25 TiO<sub>2</sub> powder [21 ( $\pm$  4.5) nm, Degussa], sodium hydroxide, and tellurium powder (200 mesh, 99.8%) were purchased from Sigma-Aldrich (Milwaukee, WI, USA). A double-beam ultraviolet–visible (UV–vis) spectrophotometer (Cintra 10e) from GBC Scientific Equipment (Dandenong, Victoria, Australia) was used to measure the absorptions of the CdTe QDs and CdHgTe NCs. High-resolution transmission electron microscopy (HR-TEM) images and energy-dispersive X-ray (EDX) spectra were recorded using an H7100 TEM system (Hitachi, Tokyo, Japan). Scanning electron microscopy (SEM) images were recorded using an ERA-8800 SEM system (Elionix, Tokyo, Japan). An X-ray powder diffraction (XRD) instrument (X'Pert PRO MRD) from PANalytical (Almelo, Netherlands) was employed to measure the crystal facets of the as-prepared nanomaterials. Current–time ( $i$ – $t$ ) plots were recorded by using a CHI 660A electrochemical analyzer (CH Instruments, Austin, TX, USA). The irradiation source for measuring the  $i$ – $t$  and photocurrent density–voltage characteristics ( $J_{sc}$ – $V$ ) was light from a 450 W xenon arc lamp (Oriol, Stratford, CT, USA) passed through an AM 1.5 filter. A Keithley 2400 digital source meter (Test Equipment Connection, Lake Mary, FL, USA) was operated under an applied external potential scan (–0.1 to +1.0 V) to record the  $J_{sc}$ – $V$  curves of the QDSSCs. The incident photon-to-current conversion efficiency (IPCE) curves were measured using a PEC-S20 instrument (Peccell Technologies, Kanagawa, Japan).

## 2.2. Synthesis of CdTe QDs

CdTe QDs were prepared according to a method described previously [19]. Briefly, sodium hydrogen telluride (NaHTe) solution was prepared by mixing tellurium powder (0.127 g) and NaBH<sub>4</sub> (2 M, 1 mL) solution. In a separate round-bottom flask, Cd(NO<sub>3</sub>)<sub>2</sub> (0.365 g), MPA solution (38 mM, pH 11.2, 74.8 mL), and the NaHTe solution (0.58 mL) were mixed and then heated at 100 °C for 20 min. The instant color change—from grayish brown to orange—indicated the formation of CdTe QDs.

## 2.3. Synthesis of CdHgTe NCs

The synthetic process for the preparation of the CdHgTe NCs was similar to that for the preparation of CdTe QDs, except that Cd(NO<sub>3</sub>)<sub>2</sub> (0.186 g) and HgCl<sub>2</sub> (0.136 g) were used instead of Cd(NO<sub>3</sub>)<sub>2</sub> (0.365 g); i.e., the total content of metal ions in the two solutions was 1 mmol. The one-pot synthesis took 50 min for the solution to change color from grayish brown to deep yellow, indicating the formation of CdHgTe NCs.

## 2.4. Fabrication of FTO–CdHgTe–FTO sandwich devices

Briefly, a mixture of CdHgTe NC solution (5 mL) and 4% PDDA (1 mL) was reacted for 10 min at ambient temperature and then it was subjected to centrifugation (3000 rpm, 10 min) to remove excess water. The resulting CdHgTe–PDDA composite was then deposited, using a scalpel, onto two clean F-doped tin oxide (FTO) conducting glass slides (resistance, 9  $\Omega$  sq<sup>–1</sup>; transmission, > 90%), each having an effective area of 2.25 cm<sup>2</sup>. After air-drying for 10 min, the two resulting glass slides were sandwiched together to develop the FTO–CdHgTe–FTO sandwich device. The  $i$ – $t$  “on/off” photoresponse of the CdHgTe NCs in this device was then recorded. Each “on/off” cycle had an interval of 10 s; the total time for five repetitive measurements was 110 s.

## 2.5. Preparation of TiO<sub>2</sub> electrodes

Cleaned FTO glass slides were used to prepare the photoelectrodes of the QDSSCs, according to a repetitive dry method [20]. TiO<sub>2</sub> paste was prepared from an aqueous solution consisting of TiO<sub>2</sub> powder (after HNO<sub>3</sub> treatment; 0.6 g), PVP (0.18 g), methyl cellulose (0.06 g), and d/d H<sub>2</sub>O (3 mL). A drop of the paste (0.08 mL) was applied to one of the bare edges of a FTO glass; it was flattened by sliding a glass rod over the tape-covered edges. A one-layer TiO<sub>2</sub> electrode was obtained after drying at 50 °C in an oven for 1 h. The process was repeated to obtain a double-layer TiO<sub>2</sub> electrode. To obtain greater values of  $\eta$ , this electrode was subjected to an additional calcination at 450 °C for 30 min. This latter coating/calcination cycle was repeated to obtain a triple-layer TiO<sub>2</sub> electrode having a thickness of 80  $\mu$ m.

## 2.6. Fabrication of TiO<sub>2</sub>–PDDA–(CdHgTe)<sub>m</sub> electrodes

The as-prepared triple-layer TiO<sub>2</sub> electrodes were used to prepare TiO<sub>2</sub>–PDDA electrodes, which are essential for improving the coating efficiency of CdHgTe NCs [18]. The TiO<sub>2</sub>–PDDA electrodes were prepared by immersion of the triple-layer TiO<sub>2</sub> electrodes, each with an effective area of 0.16 cm<sup>2</sup>, into a solution of 4% PDDA and 0.5 M NaCl for 12 h. The electrodes were then washed with d/d H<sub>2</sub>O and dried with an air gun. Each of the TiO<sub>2</sub>–PDDA electrodes was then dipped into a fresh CdHgTe NC solution in a vial. The vial was covered with a lid pierced with small holes and then it was placed in an oven and heated at 100 °C for 12 h. The electrode was removed from the vial and dried using a heat gun. The coating and drying processes were repeated up to five times to prepare yellow TiO<sub>2</sub>–PDDA–(CdHgTe)<sub>m</sub> electrodes, where  $m$  is an integer from 1 to 5. Because the TiO<sub>2</sub>–PDDA–(CdHgTe)<sub>3</sub> electrodes provided the best efficiency, they were used for further studies.

## 2.7. Fabrication of TiO<sub>2</sub>–PDDA–(CdHgTe)<sub>3</sub>–(CdTe)<sub>n</sub> and TiO<sub>2</sub>–PDDA–(CdTe)<sub>3</sub>–(CdHgTe)<sub>p</sub> electrodes

The yellow TiO<sub>2</sub>–PDDA–(CdHgTe)<sub>3</sub> electrodes were immersed into a fresh CdTe QD solution in a vial. The vial was covered with a lid pierced with small holes and then it was placed in an oven and heated at 100 °C for 6 h. The electrode was removed from the vial and dried using an air gun. The coating and drying processes—this time with a heating time of 3 h—were repeated several times to prepare TiO<sub>2</sub>–PDDA–(CdHgTe)<sub>3</sub>–(CdTe)<sub>n</sub> electrodes, where  $n$  is an integer from 1 to 4. For the preparation of TiO<sub>2</sub>–PDDA–(CdTe)<sub>3</sub>–(CdHgTe)<sub>p</sub> ( $p=1$  or 2), the coating processes were performed in the reverse order to those described in Section 2.6.

## 2.8. Fabrication of QDSSCs

All of the as-prepared photosensitized electrodes were used to prepare QDSSCs according to a previously reported method [18]. Counter electrodes were prepared by dropping an ethanol solution of Pt nanoparticles onto FTO glass substrates. After drying at 100 °C for 30 min, the Pt-coated counter electrodes were sintered at 450 °C for 30 min. Prior to use, all the as-prepared photosensitized electrodes were placed in an oven and heated at 50 °C for at least 1 h to avoid the adsorption of water. The QDSSCs were obtained after assembling one of the as-prepared photosensitized electrodes with a Pt-coated counter electrode and U-like spacer in a sandwiched configuration and annealing at 100 °C for 15 min. An EMImSCN solution containing 1.0 M LiI and 0.1 M I<sub>2</sub> was injected into each of the QDSSCs as the electrolyte.

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