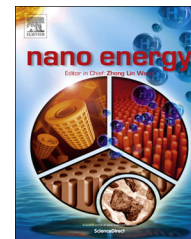




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COMMUNICATION

Hybrid tandem solar cells with depleted-heterojunction quantum dot and polymer bulk heterojunction subcells



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Abstract

We investigate hybrid tandem solar cells that rely on the combination of solution-processed depleted-heterojunction colloidal quantum dot (CQD) and bulk heterojunction polymer:fullerene subcells. The hybrid tandem solar cell is monolithically integrated and electrically connected in series with a suitable p-n recombination layer that includes metal oxides and a conjugated polyelectrolyte. We discuss the monolithic integration of the subcells, taking into account solvent interactions with underlayers and associated constraints on the tandem architecture, and show that an adequate device configuration consists of a low bandgap CQD bottom cell and a high bandgap polymer:fullerene top cell. Once we optimize the recombination layer and individual subcells, the hybrid tandem device reaches a V_{OC} of 1.3 V, approaching the sum of the individual subcell voltages. An impressive fill factor of 70% is achieved, further confirming that the subcells are efficiently connected via an appropriate recombination layer.
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Introduction

Solution-processed emerging thin film solar cells, such as devices based on organic and colloidal quantum dot (CQD) light absorbers, offer low-temperature processing, mechanical flexibility and conformability, lightweight modules, and compatibility with continuous roll-to-roll manufacturing [1-5]. Each of these is today limited to ca. 10% power conversion efficiency (PCE) in single-junction devices [6-9]. Multi-junction solar cell architectures that can harvest a broader portion of the solar spectrum are of interest to both the CQD and the organic solar cell communities [10-14]. Quantum dot solids benefit from a size-tunable bandgap that allows absorbing a relatively wide range of wavelengths from the visible to the near infrared [4,5,13,14]. Meanwhile, organic absorbers tend to have a narrower spectral absorption compared to quantum dot solids, making double (tandem) and triple-junction organic solar cells especially attractive, with published power conversion efficiency values of up to ca. 12% [15,16]. While CQD tandem solar cells have received limited attention thus far, a few CQD tandem cells have been reported with an ideal open circuit voltage (V_{OC}) equal to the sum of the two subcells (i.e. ~ 1.0 V), yet those have not shown a convincing efficiency enhancement mainly due to their modest fill factors (FF) ($< 50\%$) [13,14]. On the other hand, a key advantage of polymer tandem solar cells is their ability to achieve both high V_{OC} (i.e. > 1.5 V) and FF (i.e. $> 65\%$) [15,16]. Therefore, the opportunity exists to expand beyond the spectral range of organic absorbers and to compensate for the low FF of CQD cells by combining the two material types into a hybrid tandem solar cell which may eventually overcome the respective limitations of “all-CQD” and “all-polymer” multi-junction solar cells [17].

Here we report a hybrid tandem solar cell that combines solution-processed depleted-heterojunction CQD [18] and bulk heterojunction polymer:fullerene subcells [19,20]. The tandem is monolithically integrated and electrically connected in series with the aid of a suitable p-n recombination layer that includes metal oxides and a conjugated polyelectrolyte, and achieves high V_{OC} values and excellent FFs in the 60-70% range [21-24]. We discuss the monolithic integration of the subcells, taking into account solvent interactions with underlayers and associated constraints on the tandem architecture, and show that an adequate device configuration consists of a low bandgap CQD bottom cell and a high bandgap polymer:fullerene top cell. Once the recombination layer and individual subcells are optimized, the hybrid tandem device reaches a V_{OC} of 1.3 V, approaching the theoretical sum of the individual subcells. A FF of ca. 70% is achieved, which is higher than either of the CQD or polymer:fullerene single-junction cells, indicating that the subcells are efficiently connected via an appropriate recombination layer. Overall, the best hybrid tandem devices yield $> 5\%$ PCE, demonstrating an efficient monolithic integration of CQD and polymer:fullerene subcells, and paving the path to achieving high-efficiency hybrid tandem solar cells via appropriate solution formulations, selected interlayers, and adequate vertically-stacked configurations.

Experimental section

Single-junction depleted-heterojunction colloidal QD solar cell fabrication

Pre-patterned fluorine-tin-oxide (FTO, $15 \Omega/\text{sq}$, Xin Yan LTD.) coated glass substrates were cleaned by sonication in Extran 300 detergent solution, deionized (DI) water, acetone and isopropanol in 20 min sequential steps. Next, the substrates were blown dry by N_2 and then coated with TiO_2 nanoparticles (Dyesol) by spin-coating the diluted TiO_2 solution in ethanol (1:3 by weight) at 1500 rpm, followed by heating at 80°C for 5 min and 500°C for 30 min on a hotplate. The substrates were cooled down to room temperature, and the $TiCl_4$ treatment was performed by immersing the substrates in a 120 mM solution in DI water at 70°C for 30 min, then the substrates were rinsed with DI water and heated at 500°C for 30 min. All the procedures for the TiO_2 coating step were carried out in ambient air conditions and the substrates were stored in the N_2 glovebox after TiO_2 coating.

The quantum dot film was formed through layer-by-layer deposition of PbS CQDs (50 mg/ml in octane). For each layer, two drops (50 μl) of CQDs were cast onto the substrates through a 0.2 μm filter and spin-coated at 2500 rpm for 10 s. Next, ca. 300 μl mercaptopropionic acid (MPA) in acetonitrile (ACN) solution (1% in volume) was used to fully cover the CQD surface and kept on the surface for 3 s, the solution was then removed by spinning the sample at 2500 rpm for 5 s. Finally, the CQD surface was rinsed by casting and spinning ACN at 2500 rpm and this step was repeated twice: 5 s for the first rinse and 10 s for the second rinse. The QD deposition procedure was repeated 4 times. The PbS QD active layer has an excitonic peak at 1.29 eV (960 nm) which corresponds to an average dot diameter of 3.1 nm, based on an empirical relationship between the first absorption peak position and dot size [25]. The QD-coated substrate was then transferred to a vacuum evaporator inside the N_2 glovebox. The evaporator chamber was evacuated to 2×10^{-6} Torr for the deposition of 16 nm MoO_x (Sigma Aldrich) at a rate of 0.3 $\text{\AA}/\text{s}$, followed by 10 nm of gold (0.2 $\text{\AA}/\text{s}$), and 80 nm of Ag (0.2 $\text{\AA}/\text{s}$ for the first 10 nm and 5 $\text{\AA}/\text{s}$ for the following 70 nm) as an electrode.

Single-junction inverted bulk heterojunction polymer:fullerene solar cell fabrication

Pre-patterned indium tin oxide (ITO, $15 \Omega/\text{sq}$, Xin Yan LTD.) coated glass substrates were cleaned by the same method as that used with FTO-coated glass. For the electron transporting layer (ETL) fabrication on cleaned ITO-coated glass, ZnO powder (Sigma Aldrich) was vacuum-evaporated using a tungsten boat at a deposition rate of 0.3 $\text{\AA}/\text{s}$ in a base pressure of 6×10^{-6} Torr, followed by a conjugated polyelectrolyte, poly[(9,9-bis(3'-(*N,N*-dimethylamino)propyl)-2,7-fluorene)-alt-2,7-(9,9-dioctylfluorene)] (PFN) (1-Materials) spin-coating at 5000 rpm for 60 s. The concentration of the PFN solution in ethanol was 1 mg/ml and a small amount of acetic acid (2 $\mu\text{l}/\text{ml}$) was added. The organic

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