



Cyanine tandem and triple-junction solar cells



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ABSTRACT

Ultrathin bilayer heterojunction solar cells using cyanine electron donors and electron acceptor C₆₀ are used to fabricate monolithically stacked tandem and triple junction devices. Sub-cell stack sequences as well as C₆₀ layer thicknesses are optimized by optical modeling and maximum efficiency is corroborated experimentally. The highest power conversion efficiency of 4.3% under full sun irradiation is achieved with a tandem cell where heptamethine and trimethine cyanine dyes are used in the front and back cell, respectively. The open circuit voltage matches the sum of the two respective open circuit voltages of the individual single junction solar cells within 3%. Triple junction cells using an additional sub-cell with a pentamethine cyanine suffer from electrical series resistance. At low light irradiation intensity, however, both triple and tandem solar cells reach power conversion efficiencies above 5% in agreement with the performance increase predicted from numerical simulation.

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1. Introduction

Cyanine dyes are well known as sensitizers in silver halide photography [1], as fluorescent biomarkers [2] or as light-absorbing layers in optical data storage devices [3]. These polymethine dyes have rarely been used as solid organic semiconductors but show high promise due to very high absorption coefficients, tuneable absorption spectrum and the ability to form aggregates with highly delocalized excitons [4]. Facile and cost effective synthesis of soluble cyanines with good film forming properties add to the merits of this materials class as active semiconductors in organic photovoltaic devices (OPV) [5]. The first use of cyanine dyes as solid organic semiconductor material dates back to 1965 when Meier et al. discovered photoconductivity in thin cyanine films [6]. Much later first attempts were undertaken to use trimethine carbocyanine dyes as donors and acceptors in planar bilayer heterojunction devices [7]. Even though maximum external quantum efficiencies

of 75% were achieved [8], best power conversion efficiencies (PCE) of 3.7% in cyanine devices [9] still lag behind other small molecule semiconductors [10–12] and polymers [13–18] that recently reached PCE in excess of 8%. The main reason for this shortcoming lies in the narrow absorption band of cyanine dyes, but also in the rather low charge carrier mobility which limits the active layer thickness to 20–30 nm [19]. While the narrow width of the absorption spectrum limits photocurrent in single junction devices, well defined complementary absorption spectra are beneficial in multi-junction devices.

Organic multi-junction solar cells have been realized as a stacked assembly of single junction devices being electrically connected either in series or in parallel [20]. First multijunction solar cells were simple mechanical stacks that hardly enabled to increase the power conversion efficiency in comparison with the constituting single junction solar cells [21,22]. In some work, tandem cells were constructed out of single junctions employing the same materials which raised the efficiency due to an increased collection of photons and charge carriers [21,23]. By now, design rules are well established to optimize efficiency in monolithically stacked multi-junction organic solar cells [21,23–26]. Optical and electrical modeling are invaluable tools for hitting the maximum efficiency

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spot in the fabrication of these multilayer solar cells. When organic semiconductors with non-overlapping absorption spectra can be used, optical device optimization is particularly eased [27–29]. For example, it enables to position the low band-gap material to the front in order to optimize absorption [30]. To date, tandem [10,31] and triple-junction [32] devices with PCE exceeding 10% and 11%, respectively, have been reported. In both cases the multijunction solar cell outperformed the best constituting single junction cell by about 30%. The big challenge for further improvements is to find organic semiconductors with complementary bandgaps in the near infrared domain as well as transparent and lossless recombination layers between the monolithically stacked cells. Furthermore, solution processed cells with low energy input are of particular economic and ecologic interest [33]. Owing to their wide range of bandgaps, high extinction coefficients, solution processability and easy accessibility, cyanine dyes are interesting candidates for organic multijunction solar cells.

In previous work, we demonstrated single junction solar cells based on trimethine dyes (Cy3-P) with a band gap of ~1.8 eV [34] as well as NIR absorbing heptamethine dyes [35] (Cy7-T) with a band gap of 1.2 eV using a simple bilayer structure with C₆₀ as electron acceptor. In this work we investigate tandem solar cells with Cy3-P and Cy7-T as well as triple-junction solar cells using the latter dyes and a complementary absorbing pentamethine cyanine dye (Cy5-P). We optimized the multilayer structures by varying the thickness of the C₆₀ acceptor layers and changing the sub-cell sequence of the multijunction architecture using optical modeling based on transfer matrix formalism. Also, an efficient recombination layer composed of MoO₃ and a very thin metal layer was optimized by varying the metal and its nominal thickness. The maximum efficiency reached for tandem solar cells at AM1.5G solar irradiation conditions was 4.3% which is approximately 34% higher than the efficiency of the best single junction cell employed in the stack. Triple-junction devices were optimized in the same way as tandem cells and reached 3.6% at full solar irradiation. Tandem and triple-junction devices both reached a PCE of 5.2% at one tenth of the full sun intensity.

2. Experimental

2.1. Device fabrication

All single, tandem and triple-junction solar cells were fabricated and stored in a glove box under nitrogen (H₂O < 1 ppm, O₂ < 10 ppm). Indium tin oxide-coated glass substrates (ITO, Thin Film Devices, 140 nm, sheet resistance 20 Ohm/square) were sequentially cleaned in acetone, ethanol, detergent and de-ionized water. Layers of electron acceptor C₆₀ (SES Research, 99.5% or 99.9%), anode buffer MoO₃ (Sigma Aldrich, 99.99%), recombination metals Ag (Kurt J. Lesker, 99.99%), Al (Kurt J. Lesker, 99.999%) and Au (Kurt J. Lesker, 99.99%) as well as cathode buffer tris(8-hydroxyquinolino)aluminum (Alq₃, Sigma–Aldrich, 99.995%) were deposited by thermal evaporation under high vacuum (<5 × 10⁻⁶ mbar) at a rate of 0.1–0.3 Å/s. Active electron donor films consisting of cyanine dyes 1-ethyl-2-[3-(1-ethyl-3,3-dimethyl-1,3-dihydro-indol-2-ylidene)-propenyl]-3,3-dimethyl-3H-indolium hexafluorophosphate (Cy3-P, FEW Chemicals), 1,3,3-trimethyl-2-[5-(1,3,3-trimethyl-1,3-dihydro-indol-2-ylidene)-penta-1,3-dienyl]-3H-indolium hexafluorophosphate (Cy5-P, FEW Chemicals) and heptamethine cyanine dye 2-[2-[2-chloro-3-[2-(1-ethyl-1,3-dihydro-3,3-dimethyl-2H-indol-2-ylidene)ethylidene]-1-cyclohexen-1-yl]ethenyl]-1-ethyl-3,3-dimethyl-3H-indolium trisphate (Cy7-T, synthesized in our group) were spin coated from filtered 2,2,3,3-tetrafluoropropanol (TFP, Sigma–Aldrich) or chlorobenzene (CB, Sigma–Aldrich) solutions (spin speed of 4000 rpm

for a typical duration of 1 minute). The reason for choosing the trisphate counteranion in Cy7-T comes from the superior performance of this donor in single junction solar cells as compared to the same heptamethine dye using PF₆⁻ as a counter anion [35]. The top silver electrode (Cerac, 99.99%) was evaporated through a shadow mask to define devices with active areas of 3.1 mm² or 7.1 mm².

Bilayer single-junction devices with the general architecture ITO/MoO₃ (10 nm)/cyanine dye (10 nm or 20 nm)/C₆₀ (40 nm)/Alq₃ (2 nm)/Ag (100 nm) were fabricated by subsequent deposition steps on cleaned ITO substrates. The cyanine donor layers were deposited from solution and left for 2 h under vacuum before depositing the C₆₀ electron acceptor. TFP was used for Cy3-P and Cy5-P whereas CB was used in the case of Cy7-T. All other layers were deposited by thermal evaporation in the same vacuum chamber. Device fabrication starts with vapor deposition of MoO₃ followed by solution deposition of the cyanine layer. For the spin coating step inside a glove box, devices need to be taken out of vacuum chamber and transferred back again for vapor deposition of the remaining layers. Since the vacuum chamber is directly connected to the glove box, samples are not contaminated by ambient atmosphere during the transfer.

Tandem devices were fabricated by using ten consecutive deposition steps to form two stacked junctions with general architecture ITO/MoO₃ (10 nm)/Cy7-T (20 nm)/C₆₀ (x nm)/Alq₃ (2 nm)/RL (z nm)/MoO₃ (10 nm)/Cy3-P (10 nm)/C₆₀ (y nm)/Alq₃ (2 nm)/Ag (100 nm) named Tan 7-3 in the following. The recombination layer (RL) consisted of evaporated Au with nominal thickness z varying from 0.1 nm to 1 nm. For comparison, Ag and Al were also used. In the best performing device structure (Tan 7-3), the C₆₀ layer of the front cell had varying thicknesses x ranging from 14 nm to 35 nm, while the C₆₀ thickness of the back cell y was varied between 25 nm and 40 nm. Similarly to single junction devices, substrates have to be taken out of the vacuum chamber for solution deposition of the cyanine layers. All other layers are deposited by thermal evaporation inside the same vacuum chamber.

Triple-junction solar cells consisted of fifteen deposited layers with the general structure ITO/MoO₃ (10 nm)/Cy7-T (20 nm)/C₆₀ (x nm)/Alq₃ (2 nm)/Au (0.5 nm)/MoO₃ (10 nm)/Cy3-P or Cy5-P (10 nm)/C₆₀ (y nm)/Alq₃ (2 nm)/Au (0.5 nm)/MoO₃ (10 nm)/Cy5-P or Cy3-P (10 nm)/C₆₀ (20 nm)/Alq₃ (2 nm)/Ag (100 nm). Two promising sequences, both comprising Cy7-T as front cell were investigated. The first (Tri 7-3-5) embeds Cy3-P in the middle cell and Cy5-P in the back cell, while the second (Tri 7-5-3) incorporates Cy5-P in the middle cell and Cy3-P in the back cell. As for single and tandem devices, only the cyanine films were deposited from solution, the other layers being deposited by vapor deposition under vacuum.

2.2. Methods

For solar cell characterization, the substrates were sealed in a vacuum tight box with current feedthroughs and an optical window. Current-voltage characteristics of the solar cells were measured using a Keithley 2400 source/measure unit in the dark and under simulated AM1.5G solar irradiation of 100 mW cm⁻² from a calibrated solar simulator (Spectra-Nova). For reduced irradiation conditions, the white light intensity P_{in} was passed through neutral density filters (Andover Corporation Optical Filter). Open-circuit voltage (V_{OC}) and short-circuit current-density (J_{SC}) were obtained directly from the current–voltage characteristics. Power conversion efficiencies (PCE) were calculated as $PCE = FF \times V_{OC} \times J_{SC} \times P_{in}^{-1}$, where $FF = (J \times V)_{max} \times V_{OC}^{-1} \times J_{SC}^{-1}$ and $(J \times V)_{max}$ relates to the current-density and voltage measured at the maximum power point. A Cornerstone 130 monochromator

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