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# Series vs parallel connected organic tandem solar cells: Cell performance and impact on the design and operation of functional modules

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## ABSTRACT

Tandem solar cells are the best approach to maximize the light harvesting and adjust the overall absorption of the cell to the solar irradiance spectrum. Usually, the front and back subcells are connected in series in two-terminal device (2T) designs which require a current matching between both subcells in order to avoid potential losses. Alternatively, they can also be connected in parallel giving rise to a three terminal connection (3T). In principle, both designs have their assets and drawbacks in terms of device performance, design and materials' characterization. In this letter, we theoretically and experimentally confront both designs with each other (2T and 3T). Theoretical estimations show a maximum PCE of 15% for 2T and about 13% for 3T structures with ideal bandgaps for the front and back cell. However, 3T tandem devices can yield higher efficiencies than 2T for some specific material combinations whose theoretical values are between 10% and 12%. Therefore, other aspects related to the fabrication feasibility are studied in order to analyze the most convenient approach for module development. The need of a conducting interlayer restricts the width of the cell and causes a 3% reduction in the geometrical fill factor of the module in comparison to the 2T approach. The R2R processing of modules with 3T cells would also require an additional laser step. Finally, a couple of existing material combinations have been experimentally implemented into 2T and 3T tandem devices. The limitation imposed by their specific and non-ideal bandgaps restricts the efficiency to around 7%, considerably below the ideal case.

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## 1. Introduction

The intense research on polymer solar cells over the past decade, not only on materials but also on device performance, has taken the efficiency towards levels that were unimaginable some years ago. In this way, power conversion efficiencies over 9% are already certified [1] and the first press releases announcing values over 10% start appearing [2]. Although these efficiencies are already appealing for energy generation purposes and integration in some other applications thanks to the advanced properties that

organic semiconductors offer in comparison to their inorganic counterparts, researchers continue making persistent efforts in order to overcome the existing limitations. Absorption and thermalization losses are some of the most restricting issues at this moment of device development, since internal quantum efficiencies (IQE) over 90% and fill factors (FF) over 70% are routinely achieved by different labs [3]. This practically means that nearly all light that is absorbed is efficiently converted into current. Unfortunately, only the photons having energy higher than the bandgap of the photoactive material contribute to the energy conversion. Alike in inorganic semiconductors [4], tandem solar cells are the strategy followed to tackle this issue [5]. In spite of many efforts in this direction, polymer tandem cells do not yet show the impressive enhancement that would in theory be expected from the

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performance of optimized single cells. In this way, very recently reported works situate tandem record efficiencies over those obtained for single cells but still slightly over 10% [6,7]. The tandem approach can therefore be an interesting option for applications with area restrictions providing that there is not a substantial increase in the price per Watt-peak produced. In most of the cases, two subcells with complementary absorption are connected in series through a recombination layer where holes from one subcell meet electrons from the other. This results in a two-terminal (2T) device for which the open-circuit voltage is the summation of each subcell, and the overall current is limited by the subcell that delivers the smallest photocurrent. In an ideal situation both subcells should deliver the same photocurrent in order to minimize losses. This photocurrent matching criterion is not easy to achieve in organic solar cells. Three-terminal (3T) tandem devices are an alternative option that eliminates the need of current matching, while potential losses in terms of unbalanced photovoltages would be less dramatic for the performance of the cell [8]. In this manuscript we compare both approaches in terms of device performance, design and materials characterization. We demonstrate, experimentally and theoretically, that the 2T design is the best option to maximize the efficiency of organic tandem devices, since they benefit from higher ideally achievable efficiencies. Theoretical maximum power conversion efficiencies (PCE) are around 15% while 3T devices have the potential to reach 13%. However, the 3T design offers a larger degree of freedom in the choice of complementary absorbing materials. There are therefore some specific material combinations for which the expected efficiency for 3T devices is higher than that obtained for 2T cells. Therefore, we also analyze in detail the fabrication procedure involved for each case (2T and 3T) in order to be able to discern beforehand which design is most convenient for module development attending to the best compromise between device efficiency and level of fabrication difficulty. In addition, 3T designs also offer the possibility to independently characterize both subcells, front and back, in the same resulting structure. This would considerably ease the characterization of new materials, and would also speed up the optimization of the tandem device itself, since there is no way to check the photocurrent provided by each subcell in the 2T configuration.

#### 2. Experimental section

#### 2.1. Materials

The following materials and solvents were used as received: P3HT (American Dye Source), PTB7 (1-material), PCDTBT (Konarka Technologies),  $PC_{60}BM$  (SES Corporation) and  $PC_{70}BM$  (Solenne BV), PEDOT:PSS (Al 4083 from HC Stark), chlorobenzene (Scharlau), chloroform, ortho-dichlorobenzene (Sigma-Aldrich), and diiodooctane (Sigma-Aldrich).

#### 2.2. Solutions

The P3HT:PC<sub>60</sub>BM solution was prepared by mixing 15:12 mg in 1 mL of chlorobenzene. The solution was then heated overnight at 80  $^{\circ}$ C.

The PTB7:PC<sub>60</sub>BM was prepared in the glove box by mixing 10:15 mg in 1 mL of solvent (97% chlorobenzene and 3% diiodooctane). The solution was then heated overnight at 60 °C. Diiodooctane was added 1 h prior to the deposition of the active layer and it was left stirring at 70 °C.

The PDPP5T:PC<sub>60</sub>BM was prepared by mixing 8:16 mg in 1 mL of solvent (90% chloroform and 10% ortho-dichlorobenzene).

The PCDTBT:PC<sub>70</sub>BM was prepared by mixing 7:28 mg in 1 mL of solvent (70% ortho-dichlorobenzene and 30% chlorobenzene).

#### 2.3. Devices

ITO covered substrates were clean in subsequent acetone and isopropyl alcohol (IPA) ultrasonic baths followed by 5 min of UV ozone treatment.

#### 2.4. 2T P3HT/PTB7 series devices

Clean ITO substrates were covered with a thin layer of PEDOT: PSS deposited by spin-coating. This layer was dried at 120 °C for 10 min. The P3HT:PC<sub>60</sub>BM was deposited at different spin rates depending on the desired thickness. The P3HT:PCBM film was annealed at 140 °C for 15 min in a nitrogen filled glove box. For the intermediate layer fabrication; first, a 30 nm thick ZnO layer was spin cast at 2000 rpm from a 10 mg/ml dispersion of monodisperse 5 nm diameter crystalline ZnO nanoparticles in acetone. Details on the ZnO NP synthesis are given in [9]. Secondly, a 15 nm thick film of pH neutral PEDOT was spin cast at 2000 rpm from a 1:1 dilution of ORGACON (AGFA) and water. Next, the PTB7: PC70BM solution was spin cast at room temperature. Device fabrication was completed by thermal evaporation of 10 nm of Ca and 200 nm of Ag at room temperature under vacuum at a base pressure of  $4 \times 10^{-6}$  mbar. All devices present an active area of 9.4 mm<sup>2</sup>.

#### 2.5. 2T PCDTBT/PDPP5T series devices

On top of the dried PEDOT:PSS the PCDTBT:PC<sub>70</sub>BM was cast in nitrogen atmosphere. The resulting film was dried on a hotplate for 10 min at 70 °C. The intermediate layer was processed as before from a ZnO nanoparticles dispersion [10]. The ZnO layer was spun from a solution of 10 mg mL<sup>-1</sup> ZnO NP from isopropanol on top of the dried PCDTBT active layer. The pH neutral PEDOT:PSS (Orgacon, AGFA) was diluted 1:1 with ultrapure water after which 0.2 mL mL<sup>-1</sup> isopropanol was added to improve the wetting on the ZnO nanoparticles. The PDPP5T:PC<sub>60</sub>BM active layer was spun on top of that. Finally a back contact of 1 nm LiF and 100 nm Al was evaporated in vacuum.

### 2.6. 3T P3HT/PTB7 parallel devices

The front cell, with inverted configuration, was fabricated first. A zinc acetate (ZnAc) solution was prepared as reported by White et al. [11]. This was spin coated onto pre-cleaned ITO substrates at 4000 rpm. The resulting film was annealed at 150 °C for 5 min in order to convert the ZnAc into ZnO. The P3HT:PC<sub>60</sub>BM film was deposited by a doctor-blade with the substrate heated at 65 °C. Depending on the slot height between the substrate and the knife and the deposition speed, layers with different thicknesses were achieved. After depositing the photoactive layer, samples were covered with a Petri dish in order to create a solvent saturated environment. Following, PEDOT:PSS (Al 4083) with 1% of zonyl was spin coated on top of the P3HT:PCBM layer and the samples were annealed at 140 °C for 15 min in the glovebox. Afterwards, 10 nm of Au were thermally evaporated as interlayer. In the next step, the back cell was deposited with the regular configuration. Therefore, a layer of PEDOT:PSS (Al 4083) with 1% of zonyl was spin coated. Next, the PTB7:PC<sub>70</sub>BM layer was spin coated in the glovebox (at 1200 rpm for a 100 nm thick layer and 1700 rpm for 80 nm thick layer) followed by a drying step of 30 min at 60 °C. Finally, the top electrode was thermally evaporated, 10 nm Ca/ 150 nm Ag.

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