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Prospects of layer-split tandem cells for high-efficiency OPV

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ABSTRACT

Organic photovoltaics (OPVs) promise a low-cost alternative to traditional inorganic PV technology. Recently, the relatively poor efficiency of a bulk-heterojunction OPV has been improved considerably (> 10%) in a tandem cell configuration, albeit at the expense of using four different organic materials. In this work, we revive the concept of 'single-material tandem cells' – originally developed in the 1990s to address the transport bottleneck of a-Si solar cell. We demonstrate that OPV sub-cells made of the single polymer (or organic semiconductor) and arranged in a series tandem configuration can lead to impressive (factor of 2–5) efficiency gains, provided that the sub-cells can be thinned for optimum carrier collection and stacked for improved light absorption. Our analysis may open up a new direction in OPV research and a completely different design space for such tandem structures.

1. Introduction

Over the last 40 years, a large number of strategies have been developed to improve the efficiency of inorganic solar cells (e.g., Si, GaAs, and CdTe). These include improved light trapping, novel electrode configuration, optimum doping, effective back-surface field, passivation of grain-boundaries, suppression of Schottky barriers, tandem configuration, etc. Recently, organic photovoltaics (OPV) have emerged as a promising low-cost alternative to inorganic solar cells. Although some of the strategies translate well from inorganic thin-film technologies to OPV, the unique material properties (e.g., poor light absorption due to large bandgap, exciton bottleneck, high defect density, ultra-low mobility) suggest opportunities for OPV-specific engineering for improved performance.

The solar cell performance can be quantified by three parameters: (i) short circuit current (J_{SC}), (ii) open circuit voltage (V_{OC}), and (iii) fill-factor (FF). The optical absorption and carrier generation dictate J_{SC}. It has been known since 1960s that poor electronic properties of organic materials (i.e., poor mobility (μ) [1,2] and low recombination time (τ), summarized as the '$\mu \times \tau$' product) prevent the efficient extraction of photo-generated carriers. Thus, J_{SC} is reduced below the limit set by optical absorption. Low J_{SC} and low FF were inherent in the early OPVs where only one organic semiconductor (OS) or polymer was used as the active material [3–5]. We will denote this type of solar cells as a single-semiconductor (SS) OPV. The planer heterojunction (PHJ) OPV developed in the 1980s addressed this problem partially [6], by separating electrons and holes spatially into donor (D) and acceptor (A) semiconducting layers to minimize recombination ($\mu \times \tau$ product is improved). Eventually, this issue was addressed most successfully in 1990s when bulk-heterojuction (BHJ) OPVs were introduced, where a phase-segregated hierarchical mixture of D and A [7,8] dissociated the excitons, and carried holes and electrons to their respective contacts. The separation of transport paths for electrons and holes in the acceptors and donors, respectively, ensured improved extraction of photo-generated carriers [9]. The improvement in J_{SC} was somewhat counterbalanced by a reduction in V_{OC}, because the V_{OC} of BHJ-OPVs is limited by the cross-gap of the band-discontinuity (Δ_{DA}) created at the D–A interface [9,10,38]. Overall, the performance is improved by using a tandem configuration, where a second BHJ-OPV sub-cell (based on two new D and A) is stacked onto the first BHJ-OPV. Indeed, highly efficient cells (> 10%) has been developed based on these schemes. Unfortunately, process integration of various materials has always been a concern for tandem configuration [11–13]. The issue is somewhat more complicated for BHJ-OPV, where each sub-cell consists of two organic semiconductors (i.e., n subcells require $2n$ organic semiconductors). Moreover, the continuous phase segregation of D and A materials makes reliability and variability important issues for BHJ-OPV [9,14,15].

An approach to recover V_{OC} is to reconsider the prospect of a SS-OPV, so that the cross-gap penalty intrinsic to BHJ-OPV is avoided. Moreover, the intrinsic reliability issues associated with D–A phase segregation are presumed absent in SS-OPV. Indeed such SS-OPV has been recently experimentally demonstrated [16]. This strategy of using a single absorber layer is typical for many thin-film technologies, such as a-Si, CdTe, and CIGS. Unfortunately, for an absorber of thickness L, the very low mobility of OS creates a trade-off between absorption (thicker is better) and transport (thinner is desired). Specifically, OS are inherently p-type or n-type [16–18]. Hence, one

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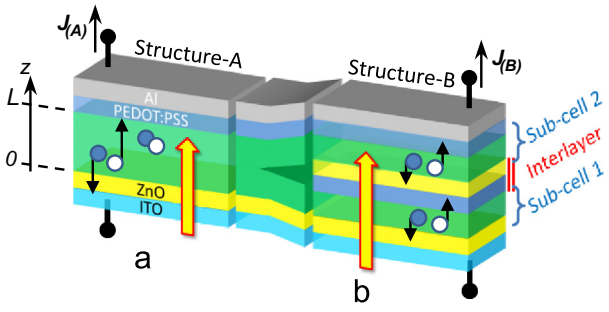

Fig. 1. (a) A single active layer PV referred to as structure-A. (b) Tandem PV consisting of sub-cells constructed from the same active layer material (structure-B). The strategy involves splitting the thick active layer of structure-A to form the thinner sub-cells of the tandem structure-B. We define the *z*-axis to be through the depth of the active layer. The origin of this axis is at the interface of the electron collecting (ZnO) layer and the active layer.

of the contacts (either electron collecting or hole collecting) forms a Schottky junction [16], or space charge layer (SCL). The electric field within the SCL ($L_{SCL}\sim$50 nm, at doping-density $\sim 10^{17}$ cm^{-3}) allows efficient charge collection, but the poor mobility dictates that a large fraction of the charge generated within the field-free region and a diffusion length (L_D) away from the junction is lost to recombination [39]. Ideally, efficient charge collection requires $L\sim L_{col}\equiv(L_{SCL}+L_D)$, however, very poor absorptance of such a thin layer degrades J_{SC}. For a rigorous definition of L_{col}, see Section S1 in the Supporting information (SI).

In this paper, we study the 'layer-split-tandem' structure for the single OS to overcome the absorption vs. transport trade-off related to SS-OPV. The concept was originally introduced in the 1990s to address charge-transport bottleneck in a-Si PV [19–21] – a material with relatively poor mobility ($\mu_{a-Si}\sim 20$ cm^{2}V^{-1} s^{-1}) compared to other inorganic semiconductors. Since $\mu_{OS}\sim 10^{-5}$–10^{-2} cm^{2}V^{-1} s^{-1}, the concept of 'layer-split-tandem' could offer significant improvement. Indeed, the validity of the idea has been already demonstrated – albeit with BHJ-OPV [22–24] defined by two OS.

Briefly, the idea involves splitting a thick OPV layer (L) into multiple sub-cells (thicknesses L_1, L_2,…) connected is a series tandem configuration, see Fig. 1. The current in each subcell must be matched for maximum efficiency (i.e., $I_1=I_2=\ldots I_i$). The overall stacked cell is optically thick with good light absorption, while the individual sub-cells are electrically thin to ensure good carrier extraction. Moreover, the use of single OS offers potential for high V_{OC} (and hence higher efficiency) compared to BHJ. We explore the relative merits of this single-polymer strategy for OS characterized by a wide range of mobilities and absorption coefficients. We find that even a two-cell tandem offers considerable improvement (factor of $\sim$2), and we establish the number of sub-cells (N) that would maximize the efficiency. Remarkably, the ultimate efficiency may approach the SQ limit associated with these devices, provided the interlayer absorption (ILA) is neglected. In practice, ILA limits the number of sub-cells that can be integrated without sacrificing overall efficiency.

The paper is arranged as follows. We describe the concept qualitatively in Section 2 and explain the numerical modeling framework in Section 3. We summarize the results for SS-OPV, 2-layer and N-layer tandem OPV in Section 4. The key conclusions are highlighted in Section 5.

2. The concept of a layer-split-tandem cell

The structures shown in Fig. 1(a and b) explain the concept of splitting an active layer to form a tandem cell. Structure-A corresponds to a solar cell with a thick active layer which can absorb almost all the photons (with energies above the OS band-gap) incident onto the structure. This is the traditional SS-OPV we mentioned in the Introduction. On the other hand, structure-B has the layer-split-tandem configuration with the same total absorption as in structure-A, and optically designed to ensure current matching between the two split layers. In other words, photo-generated carrier densities of both the structures are identical. For simplicity, let us assume that the total absorption (GL) in structure-A to be equally split ($GL/2$) into the 2 sub-cells of structure-B. Hence, the photo-generated carrier density G remains the same from structure-A to the sub-cells of structure-B.

Now consider the charge-collection efficiencies of structure-A vs. structure-B, *assuming* the idealized case that the carrier mobilities are high, the recombination rates are low, and interlayer absorption (ILA) and series-resistance ($R_{s(int)}$) are negligible. For structure-B, a perfect optical design will yield half the absorption in each split-layer sub-cell compared to that of structure-A, so that $J_{SC(B-ideal)}=J_{SC(A-ideal)}/2$, in the limit of high mobility and efficient charge extraction. The series tandem configuration yields $V_{OC(B)}=2V_{OC(A)}$. The *FF*s remain approximately the same. Thus, the efficiency $\eta_B=J_{SC(B-ideal)}V_{OC(B)}FF=J_{SC(A-ideal)}V_{OC(A)}FF=\eta_A$ remains unchanged. Therefore, *the layer-split tandem configuration provides no advantage for high-mobility material*, explaining why this concept is irrelevant for traditional inorganic semiconductors.

The aforementioned conclusion changes dramatically if the carrier mobility is low and the recombination rates are high, as is the case for OS or polymer. Also, the material being inherently p or n-type, the photo-generated carriers within the depletion region (L_{col}) near the Schottky junction can be collected (See Fig. 2(a)) by the electric field and the photo-generated carriers in the field free region will recombine before being collected by the contacts. Therefore, $J_{SC(A)}\sim GL_{col}$ and while $J_{SC(B)}\sim GL_{col}$ (in each sub-cell), assuming same photo-generated carrier density G for structures A and B. In other words, $J_{SC(A)}\sim J_{SC(B)}$. Moreover, improved charge collection ensures that $FF_{(B)}>FF_{(A)}$. The reduced dark-current in structure-B ensures that $V_{OC(B)sub-cell}\geq V_{OC(A)}$ translating into $V_{OC(B)}\geq 2V_{OC(A)}$, and therefore, $\eta_B>2\eta_A$ – *the split-tandem could be more than twice as efficient as its single junction counterpart*. Indeed, so long the $L_{col}<L_i$, the thickness of the thinnest sub-cell, the cell efficiency should increase linearly with the number of sub-cells. Obviously, the improvement will be reduced once ILA and interlayer series resistance $R_{S(int)}$-loss are accounted for. Conceptually, therefore, SS-tandem appears as a

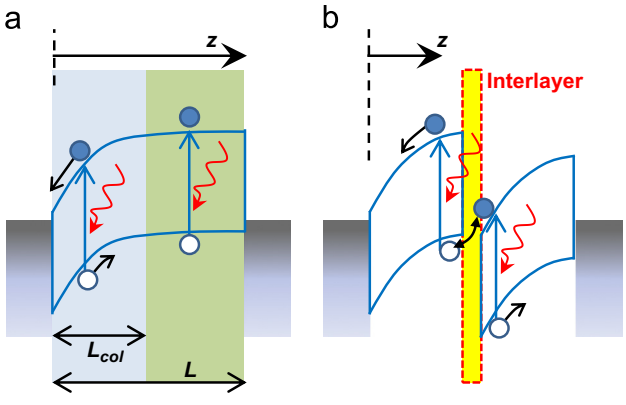

Fig. 2. (a) Schematic band diagram for a p-type organic semiconductor, showing band bending due to formation of Schottky contact (left contact). Only carriers from the L_{col} region can be collected. Carriers from the quasi neutral region (shaded dark) are lost to self-recombination. (b) A schematic band diagram for the split-tandem solar cell. Each of the sub-cells is characterized by their respective Schottky junction. The interlayer (yellow) provides a low resistance path for electron–hole recombination. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)