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### Solar Energy Materials & Solar Cells



# Efficient hybrid inorganic/organic tandem solar cells with tailored recombination contacts



Solar Energy Material

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#### ABSTRACT

In this work, the authors present a 7.5% efficient hybrid tandem solar cell with the bottom cell made of amorphous silicon and a Si-PCPDTBT:PC<sub>70</sub>BM bulk heterojunction top cell. Loss-free recombination contacts were realized by combing Al-doped ZnO with either the conducting polymer composite PEDOT: PSS or with a bilayer of ultrathin Al and MoO<sub>3</sub>. Optimization of these contacts results in tandem cells with high fill factors of 70% and an open circuit voltage close to the sum of those of the sub-cells. This is the best efficiency reported for this type of hybrid tandem cell so far. Optical and electrical device modeling suggests that the efficiency can be increased to ~12% on combining a donor polymer with suitable absorption onset with PCBM. We also describe proof-of-principle studies employing light trapping in hybrid tandem solar cells, suggesting that this device architecture has the potential to achieve efficiencies well above 12%.

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#### 1. Introduction

The last decade was shaped by a tremendous increase in efficiency of organic solar cells. Both polymeric and small molecular materials processed either from solution or by vacuum deposition reached commercialization acceptable efficiencies around 10% in single junction [1,2] or tandem cell configuration [3,4]. This improvement shows the great potential of organic solar cells to be implemented into a low production and installation cost device technology [5]. One of the key advantages of organic materials is the overall high absorptivity and the sharp absorption onset. Thus, very thin organic films of approximately 100 nm thickness can be utilized to absorb almost 80% of the incident sunlight in the active organic material within a broad range of wavelengths in a device structure having the reflecting back electrode close to the active layer [6]. However, in the tandem cell configuration, the wide bandgap front cell should be in the range of 200-300 nm thickness for optimum light absorption in the case of organic front cells [3]. Additionally it needs to have a high fill factor (FF) and open circuit voltage  $(V_{oc})$  at this optically optimized thickness. A general issue for efficient organic materials is a significant extraction loss if the

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http://dx.doi.org/10.1016/j.solmat.2014.04.020 0927-0248/© 2014 Elsevier B.V. All rights reserved. active layer exceeds 100 nm [7–9]. This results in lower FFs when used as tandem front cells [10]. P3HT:ICBA is the material combination that works the best so far as front cell in organic multijunctions with appropriate thickness [3], although this blend is yet not optimized in terms of internal quantum efficiency and the optical band-gap versus open circuit voltage relation. There are other organic material combinations with comparable optical absorption onsets showing either higher  $V_{\rm oc}$  with lacking FF at thicknesses above 200 nm [11], or a higher FF at thick layers but a smaller  $V_{\rm oc}$  [12]. At the moment, the most promising wide band-gap organic material suitable for the front cell in a tandem configuration is the PBDTFBZS:PC<sub>70</sub>BM blend, reaching 7.7% efficiency in single junctions with 250 nm layer thickness [13].

Amorphous hydrogenated silicon (a-Si:H) has a band-gap of 1.7 eV and shows a  $V_{\rm oc}$  of ~0.9 V together with internal quantum efficiencies of 100% (in the i-layer) and FFs of 70%, even for the thickness range of 100–250 nm. In optimized laboratory scale single junctions the highest reported stabilized efficiency for a-Si:H is about 10% with a ~250 nm (i) utilizing light trapping [14]. This is superior to P3HT:ICBA or other wide band-gap organic single junction cells that have been employed in tandem devices so far. a-Si:H is often used in tandem structures in combination with microcrystalline Si:H ( $\mu$ c-Si:H). The disadvantage of  $\mu$ c-Si:H is the low absorption in the red to NIR spectra as seen in Fig. 1. The hybrid tandem cell configuration, made from an a-Si:H front cell

and an organic back cell therefore, combines the advantages of both the inorganic a-Si:H front cell and the organic low-band-gap back cell in one device.

Here we investigate hybrid tandem cells with a thin ( $\sim$  140 nm) a-Si:H front cell connected in series with an organic bulk heterojunction sub-cell made from the low band-gap donor polymer Si-PCPDTBT blended with PC<sub>70</sub>BM. This hybrid device design is a quite new approach. In the last two years, only a few reports have been published using this hybrid design, with efficiencies of 1.8-3.3% [15–17] and 5.7% [18] as reported by Kim et al. and Seo et al., respectively. Kim and coworkers use a low performing organic sub-cell made of PCPDTBT:PCBM, but without using processing additives which are necessary to get good solar cell performance [19]. The work published by Seo and coworkers makes use of the polymer PBDTTT-C having a rather high band-gap of  $\sim$  1.63 eV. To generate high currents in the organic sub-cell in combination with a-Si:H, the band-gap needs to be lowered further. Importantly, both papers show tandem cells with FFs below 50%, highlighting the importance of using appropriate recombination contacts in



**Fig. 1.** (Left scale) Extinction coefficient k together with (right scale) the spectral photon flux density of AM 1.5G spectra.

this hybrid configuration. Also, both reports make use of indium tin oxide (ITO) as transparent conductive oxide which is generally avoided for a-Si:H solar cells.

By using organic sub-cells with high quantum efficiencies up to 800 nm and optimized recombination contacts together with optimizing light management, we reach a power conversion efficiency (PCE) of 7.5% in a hybrid tandem cell made from a-Si: H and Si-PCPDTBT:PC<sub>70</sub>BM sub-cells. We present guidelines for further efficiency enhancements by tuning the polymer band-gap in the organic blend, with predicted hybrid tandem cell efficiencies close to 12%. Additionally, we show proof-of-concept studies for light trapping by textured front contacts. This strategy could further enhance the efficiency well above 12% when properly implemented in hybrid tandem solar cells.

Fig. 1 shows the absorption strength of a- and  $\mu$ c-Si:H together with the organic P3HT:IC<sub>60</sub>BA and Si-PCPDTBT:PC<sub>70</sub>BM blends. This comparison displays the strong absorption onsets of the organic materials compared over Si:H, especially in the range above k=0.01. Fig. 1 also displays the complementary absorption between a-Si:H and Si-PCPDTBT:PC<sub>70</sub>BM, and the wide spectral range covered by these two materials.

#### 2. Results

One of the standard transparent conductive oxides (TCOs) used for a-Si:H or  $\mu$ c-Si:H cells is aluminum doped zinc oxide (AZO) [20–22]. AZO can be easily sputtered with smooth surfaces and etched with e.g. diluted HCl to texturize the front contact and generate light trapping effects [20,23,24]. Tin doped indium oxide (ITO) which is usually used for organic electronics suffers from the reduction by hydrogen radicals during a-Si:H or  $\mu$ c-Si:H layer deposition resulting in reduced transparency when applied as front contact [25]. We, therefore, built our hybrid layer stack on AZO as the front contact as displayed in Fig. 2. The 700 nm thick front AZO is deposited via sputtering. On top of that, a layer sequence of p-type, intrinsic and n-type a-Si:H (together ~ 140 nm) is deposited via plasma enhanced chemical vapor deposition (PECVD). Finally,



**Fig. 2.** (a) Schematic device structure with a  $Ag/MoO_3$  recombination layer and (b) device structure with PEDOT:PSS as the p-type recombination contact. (c) J-V characteristics of devices with the structure shown in (a). (d) J-V characteristics of devices from (b). All J-V characteristics shown here are measured under AM 1.5G at 100 mW/cm<sup>2</sup> without the use of an aperture.

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