

Scientific Report

about

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1 Introduction

Within the last decade polymer solar cells – as part of organic electronics – have undergone a tremendous development in terms of understanding principal device function and improving power conversion efficiency. By surpassing certified efficiencies of 7% (Solarmer, CA) within the last year, the polymer solar cell concept has reached application levels and is on the way to commercialization. One key for further enhancing the power conversion efficiency beyond 10% is offered by the concept of tandem solar cells. Originally introduced for small molecule based evaporated solar cells [1-4], the concept has been extended to solution based solar cells recently [5-9].

As shown in the case of small organic molecular solar cells [1, 2], an interstitial gold or silver layer functioned as charge collection and recombination layer between the two sub-cells of the tandem solar cell. In the case of solution based tandem solar cells, mostly more complicated interlayers were used – often based on solution processible metal-oxides [6, 8]. However, one further aspect for using metal nanocluster-based recombination layers is a potential increase in absorption of the solar cells due to plasmonic effects [10].

The main purpose of this scientific collaboration was to investigate whether a simple evaporated metal-based interlayer can be applied for the intermediate recombination layer. In a series of initial experiments by my student Annemiek Chall, only employing calcium, gold or silver for the interlayer, did fail due to dissolution of the firstly deposited sub-cell by the second. Hence we aimed for a better stability of the recombination interlayer by addition of PEDOT:PSS on top of the metal clusters. Further we tried to apply even insulating polymers for better stabilization against dissolution. For proper tandem cell operation, a lower bandgap polymer – named PS41 – (synthesized by Dr. Raja Shahid Ashraf) was brought with me for further use.

2 Experimental

2.1 Device Preparation

Organic semiconductor thin film devices studied herein were prepared in sandwich geometry (see Figure 1). Substrates for these devices are 1.5 x 1.5 cm large glass sheets covered with Indium-Tin-Oxide (ITO). To ensure secure connection of the metal top electrode without the risk of punching through the metal layer and shortcut to the ITO, part of the ITO was removed by chemical wet etching. To protect the part of the substrate from acid where the ITO-layer should remain, it was coated with a sticky tape. For thorough cleaning substrates subsequently underwent 15 minutes in acetone and isopropanol supersonic bath. After drying, the substrates were covered first with PEDOT:PSS and then with the organic semiconductor layer by spin coating. As next a metallic interlayer was evaporated from gold or silver. On top of this, a second organic semiconductor layer was spin cast and the device was finalized by evaporation of an aluminium back electrode.

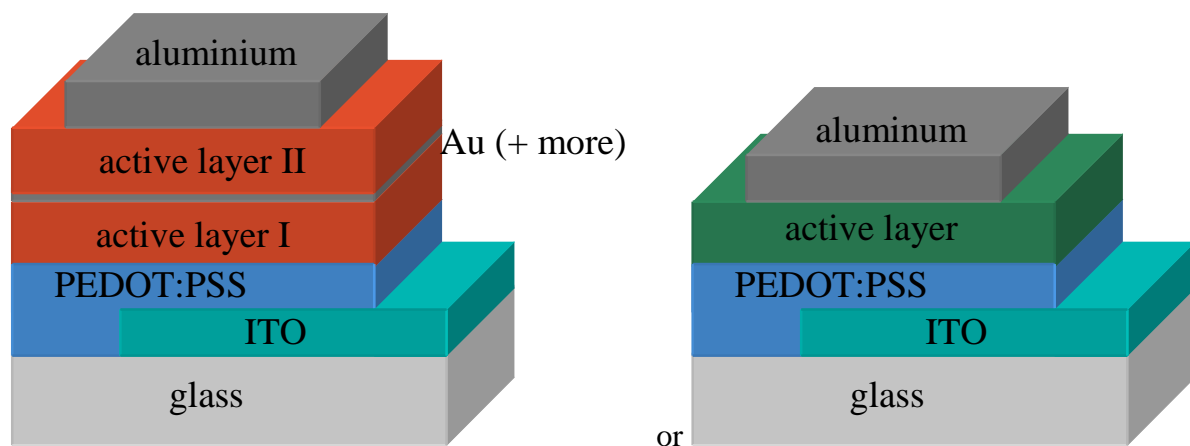
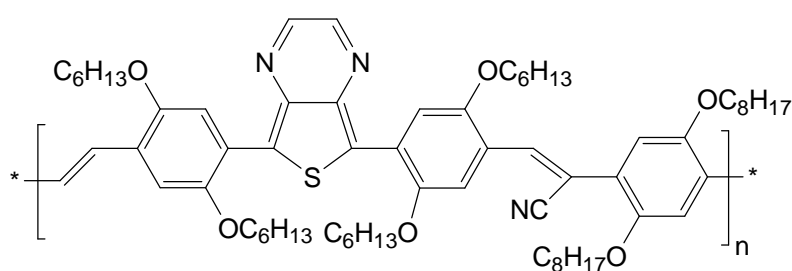


Figure 1: Design for a tandem organic solar cell (left) and simple single solar cell (right).

Active layers in the case of tandem solar cells were prepared from regio-regular poly(3-hexylthiophene-2,5-diyl) (P3HT, purchased from Rieke Metals, Inc.) and (1-(3-methoxycarbonyl)propyl-1-phenyl [6,6]C 61) (PCBM, purchased from Solenne) by spin coating from a solution in chlorobenzene (CB). The single solar cells were prepared from PS41, depicted in Figure 2.



PS-41

Fig. 2: Chemical structure of the low bandgap polymer PS41, kindly provided by Dr. Raja Shahid Ashraf.

For solar cell preparation the following sample sets were done:

1.) Low bandgap single solar cells from PS 41:PCBM blends (1:1 by weight)

PS41 spin freq.	700 s⁻¹	600 s⁻¹	500 s⁻¹	400 s⁻¹
Not annealed	A171	A174	A177	A180
120°C (1 min)	A172	A175	A178	A181
150°C (1 min)	A173	A176	A179	A182

2.) Tandem solar cells with P3HT:PCBM blends (1:1 by weight):

The intermediate layer was either made of 5 nm evaporated gold, 5 nm gold with additional PEDOT:PSS layer or a combination of first 2.5 nm calcium with 5 nm gold on top. Further variations were done by spin casting PMMA on top of the Au or Ca/Au electrodes and by spin casting PEDOT:PSS on top of the Au/PMMA bilayer.

Coating of electrode	Au 5nm	Au5nm/PEDOT:PSS	Ca2.5nm/Au2.5nm
nothing	A159, A160	A161, A162	A163, A164, A165
PMMA	A151, A158, A154	---	A166, A167, A168, A170
PMMA/PEDOT:PSS	A157, A155, A153	---	---

The sample numbers in red are for P3HT:PCBM stacked solar cells alone (bottom and top cell are the same) and the sample numbers in green stand for tandem solar cells with P3HT:PCBM bottom and PS41:PCBM top cells.

Spin frequencies used were 1000 rpm for P3HT:PCBM active layers and 400 rpm for PS41:PCBM active layers. Annealing of the tandem solar cells was done at 120°C for 2 min.

2.2 Experimental Methods

After device preparation the light I-V-characteristics under AM 1.5 solar irradiation simulation of all organic solar cells were determined by a Keithley source-measure-unit under inert conditions.

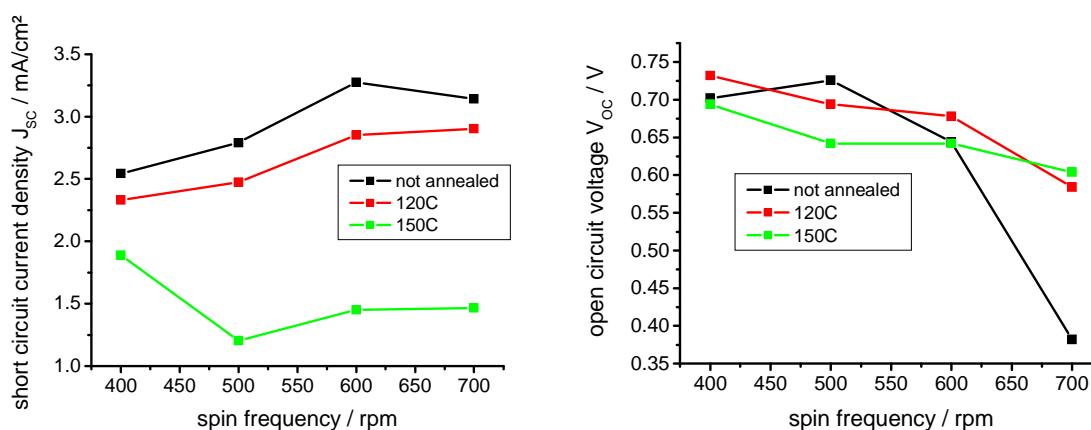
To determine the thickness of the PMMA-interlayer, films spin cast on glass were scratched and film thickness was measured by use of a tactile profilometer.

3 Results

In the following a brief description of the main results is given. In the first section single solar cells based on PS 41 are reported, while in the second section results for stacked and tandem solar cells are presented. Unfortunately no properly working tandem solar cells could be achieved, since the dissolution problem of the lower sub-cells remained even when using PEDOT:PSS and/or PMMA as protection for the interlayer.

3.1 PS 41 based single solar cells

Single solar cells based on PS 41:PCBM blends yielded in the first round of optimization up to about 0.5% efficient photovoltaic devices. The optimization based on a variation of spin frequencies and annealing conditions is depicted in Fig. 3.



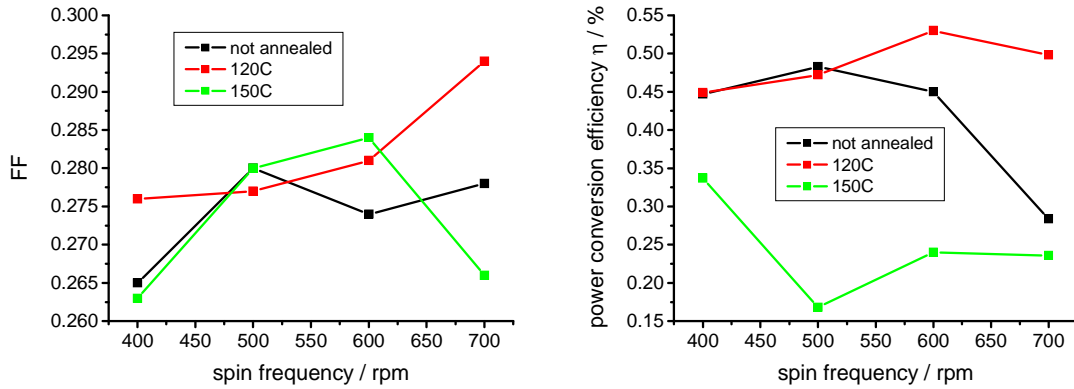


Figure 2: Optimization of solar cells based on PS 41:PCBM blends by variation of spin frequency and annealing conditions. The maximum power conversion efficiency of 0.53% was achieved for mild annealing conditions of 1 min at 120°C.

The optimization yielded 0.53% power conversion efficiency at the maximum for solar cells under mild annealing conditions of 120°C for one minute. Further increase in annealing temperature led to fatal loss in short circuit current density. Thus the solar cells are operating on a rather low overall performance, controlled by small short circuit currents and very low fill factors.

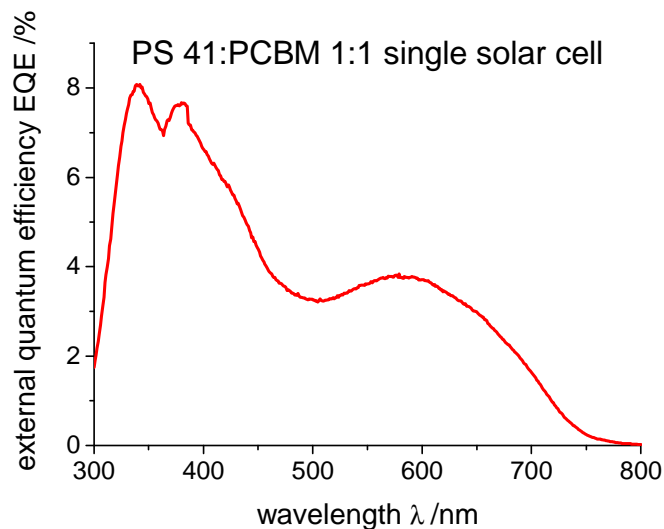


Figure 4: External quantum efficiency spectra for a PS 41:PCBM blend solar cell. In fact the absolute values given here seem to underestimate the photocurrent densities shown in Fig. 3.

From the external quantum efficiency spectrum it can be understood that the absorption extends further into the red part of the solar spectrum, however, the gain as compared to P3HT (not shown here) is in the range of about 100 nm only.

In conclusion, tandem solar cells together with P3HT-based solar cells could be suitable for demonstration purposes only.

3.2 P3HT-based tandem solar cells with various interlayers

Most of the stacked and tandem solar cell devices suffered severely from shunting behaviour, probably caused by the dissolution of the gold interstitial layer and the bottom cell. Thus in general the device efficiencies remained poor. As an example for the best operating devices samples A163 (ITO/PEDOT:PSS/P3HT:PCBM/Ca/Au/P3HT:PCBM/Al) and A165 (ITO/PEDOT:PSS/P3HT:PCBM/Ca/Au/PS 41:PCBM/Al) are compared in Fig. 5. As can be seen from the IV-characteristics, the open circuit voltage V_{OC} did not exceed the expectable value (~ 0.6 Volts) for P3HT:PCBM based solar cells at any time. Thus, no tandem operation could not be identified in these devices.

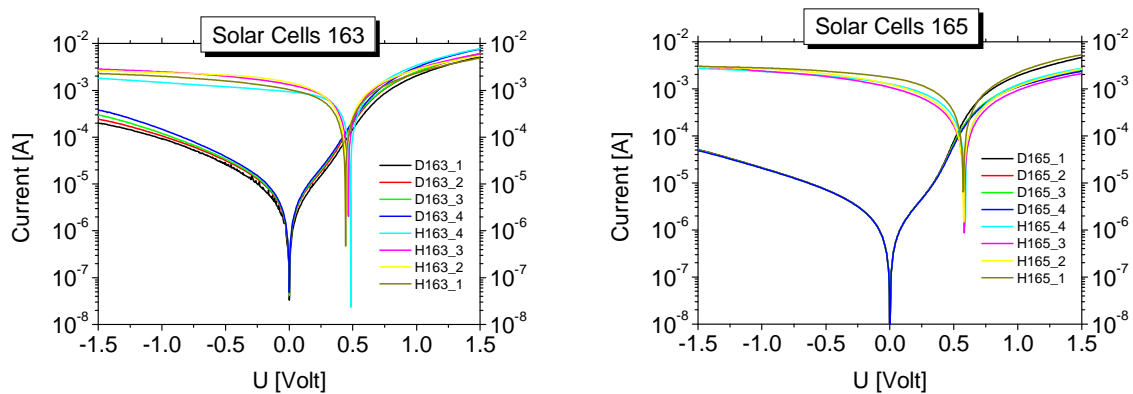


Figure 5: Dark and light I-V-characteristics for stacked solar cells A163 (only P3HT-based) and A165 (with P3HT-based bottom cell and PS 41-based top cell) are shown. As interstitial layer a sequence of Ca and Au (each 2.5 nm) was used.

The addition of PMMA did not brought about any improvement in terms of open circuit voltage – in agreement with the results for the pristine Au-interstitial layer. The PMMA films were spin cast from a different solvent system so that their deposition did not affect the gold interlayer as such. With the help of a tactile profilometer, PMMA film thicknesses of about 10 nm were projected in the tandem devices.

In general, no voltage doubling was achieved for any device and hence the recombination layers were defected in all cases. This fact has been related by some microscopy images, from which the dissolution of the underlying P3HT:PCBM sub-cell could be made visible (not shown here).

In conclusion, all of the prepared tandem and stacked solar cell behaved in a similar way. No interlayer structure brought about specific advantages over the others, since all devices suffered from dissolution of the interlayer and the underlying bottom sub-cell. Hence more solvent-stable interstitial layers need to be used for tandem solar cell operation.

4 Future collaboration and projected publications

At the moment no detailed work plan exists between Prof. Wantz and me for further collaboration. I have started to apply TiO_x-based interstitial layers for the preparation of tandem solar cells in Ilmenau. In case these experiments yield successful results, a reactivation of the collaboration seems feasible.

5 References

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