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Simple, Highly Efficient Vacuum-Processed Bulk Heterojunction Solar Cells Based on Merocyanine Dyes

Vera Steinmann, Nils M. Kronenberg, Martin R. Lenze, Steven M. Graf, Dirk Hertel, Klaus Meerholz,* Hannah Bürckstümmer, Elena V. Tulyakova, and Frank Würthner*

In order to be competitive on the energy market, organic solar cells with higher efficiency are needed. To date, polymer solar cells have retained the lead with efficiencies of up to 8%. However, research on small molecule solar cells has been catching up throughout recent years and is showing similar efficiencies, however, only for more sophisticated multilayer device configurations. In this work, a simple, highly efficient, vacuum-processed small molecule solar cell based on merocyanine dyes – traditional colorants that can easily be mass-produced and purified – is presented. In the past, merocyanines have been successfully introduced in solution-processed as well as vacuum-processed devices, demonstrating efficiencies up to 4.9%. Here, further optimization of devices is achieved while keeping the same simple layer stack, ultimately leading to efficiencies beyond the 6% mark. In addition, physical properties such as the charge carrier transport and the cell performance under various light intensities are addressed.

1. Introduction

Organic solar cells (OSCs) have attracted considerable interest due to their potential for large-scale, cost-effective and environmentally friendly power generation. During recent years, OSC research based on the bulk heterojunction (BHJ) concept has shown substantial progress, reporting power conversion efficiencies (*PCE*) beyond 5% for vacuum-deposited (VAC) small-molecule BHJ cells^[1–3] and even higher *PCEs* (up to 7.4%) for solution-processed (SOL) polymer-based BHJ devices.^[4,5] In OSCs, small molecules (SMs) are of increasing interest due to their outstandingly high absorption coefficients and simple purification procedures in contrast to commonly

used conjugated donor polymers, such as poly(3-hexylthiophene) (P3HT) or the more recent donor–acceptor-type polymers (e.g., PCPDTBT).

Forrest et al. reported on a VAC device containing four organic layers (ITO/CuPc/CuPc:C₆₀/C₆₀/BCP/Ag) with $(5.0 \pm 0.3)\%$ efficiency. In recent years, prospering research has led to an increased number of publications on highly efficient SM solar cells. However, in order to improve *PCE* rather complicated tandem solar cell structures have been realized. In 2004, again Forrest et al. presented a tandem cell with ten VAC-processed layers between the electrodes leading to an impressive *PCE* of $(5.7 \pm 0.3)\%$.^[2] Five years later, Bruder et al. succeeded in further increasing the *PCE* to $(6.0 \pm 0.1)\%$ by introducing a combination of a solid-state dye-sensitized cell and a

VAC-deposited BHJ solar cell.^[6] This tandem device consisted of seven different layers. The exact structure of recent record tandem-devices reported by Heliatic is unknown, however, it is safe to assume that it comprised more layers than the corresponding single cells (i.e., not tandem, see Fitzner et al.^[3]).

Despite this success, it is obvious that even higher *PCEs* and much simpler cell structures are inevitably required to make OSCs cost-effective. Therefore, the development of SMs with high absorption coefficients and favorable film morphologies is needed. Very recently, the Leo and Bäuerle groups presented a single-cell SM-BHJ solar cell based on dicyanovinyl-substituted oligothiophenes with a high *PCE* of up to 5.2%.^[3] To the best of our knowledge, this efficiency has been the highest value reported for single VAC-processed SM-BHJ solar cells. However, the device setup still comprises seven organic layers, some of which are redox-chemically doped (*pin*-concept).

In this publication, we present a highly efficient VAC-processed single-cell SM-BHJ solar cell based on merocyanine (MC) dyes, surpassing the 6% mark. We have chosen a remarkably simple layer stack that comprises only two organic layers. Thus, in comparison to the single-cell oligothiophene-based OSC,^[3] we are able to demonstrate an additional efficiency improvement by almost 20%, while at the same time reducing the layer stack from six to only two additional layers without the need for redoxchemical doping.

MC dyes are small molecules characterized by a rather strong dipolar character and high polarizabilities, enabling high

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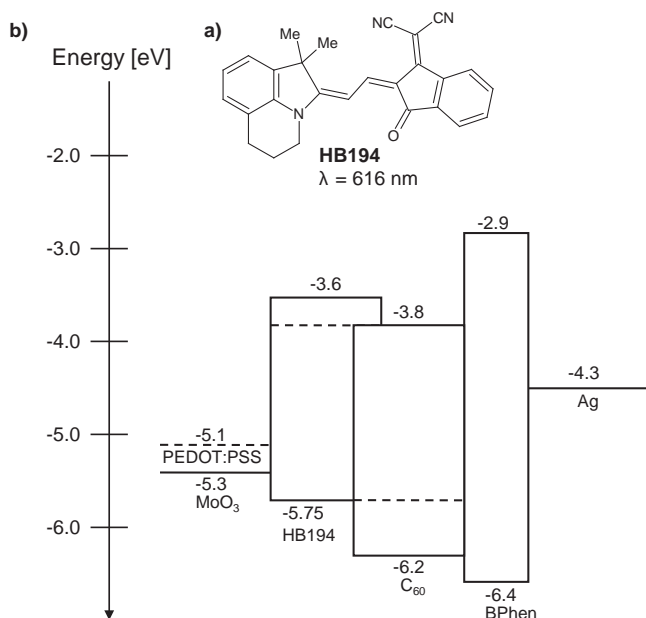


Figure 1. a) Chemical structure of the MC dye HB194 with its maximum absorption wavelength obtained from thin-film UV/Vis-measurements. b) The schematic energy alignment shows PEDOT:PSS and MoO₃ as HCC, the MC dye HB194 as electron donor and C₆₀ as electron acceptor in a BHJ structure, followed by BPhen as EBL, and Ag as top electrode.^[10,12,13,31–33]

absorption coefficients.^[7–9] In our previous work, MC dyes have been subject to detailed studies and have been successfully modified in order to achieve highly efficient small dye molecules for SOL-processed OSCs.^[9,10] For the most efficient MC dye (HB194) among our SOL-processed devices we have subsequently also prepared VAC devices that exhibited a *PCE* of 4.9% before cell optimization and using a notably simple layer stack consisting of only three organic layers.^[11]

2. Results and Discussion

In this work, we report the further optimization of a VAC-processed single SM-BHJ solar cell based on the MC dye HB194 (see Figure 1a for the chemical structure) while sticking to an extremely simple device setup, which is favorable for production purposes. The presented solar cell stack consists of an active BHJ layer sandwiched between a hole-collecting contact (HCC) on the one side and an exciton-blocking layer (EBL) on the other. In all devices, we have used a thin layer of 4,7-diphenyl-1,10-phenanthroline (BPhen) on top of the active layer, followed by a silver (Ag) electrode. In the literature, BPhen is widely used as an EBL due to its large energy gap of 3.5 eV.^[12] The material is highly transparent and hence does not contribute to the light absorption of the cell. The optimum BPhen thickness was determined to be 6 ± 2 nm. For thicker EBLs we observed an efficiency decrease which could result from an increasing series resistance (*R_s*) in thicker BPhen layers.^[12] The presented solar cell stack was entirely processed under high-vacuum conditions in a custom-made chamber (K. J. Lesker Co., UK).

In our previous work, we used PEDOT:PSS as the HCC.^[9–11] For reasons such as improving processing conditions, we intended to substitute the SOL-processed PEDOT:PSS by a VAC-processed HCC. Over recent years, MoO₃ has become a common candidate for an HCC in VAC-processed OSCs. Recently, there has been a controversy in the literature about the role of transition metal oxides such as MoO₃ in charge-generation layers. Ultraviolet photoemission spectroscopy (UPS) measurements by Yang et al. showed MoO₃ being a p-type semiconductor with a valence band at -5.3 eV.^[13] Data presented by Forrest et al. supported this view.^[14] However, recent publications by Kahn et al. suggest a very deep lying conducting band at -6.7 eV for MoO₃ and a high work function of -6.9 eV.^[15–17] According to this model MoO₃ acts as a n-type semiconductor.

Irrespective of this controversy, we tested MoO₃ as an HCC in our devices. It turned out that replacing PEDOT:PSS with MoO₃ resulted in a significant improvement in the device performance under standard AM1.5 illumination conditions (100 mW cm^{-2}). We observed a remarkably high open-circuit voltage (*V_{OC}*) of up to 970 mV for MoO₃ cells (device B), whereas PEDOT:PSS cells only reached a *V_{OC}* of 770 mV (device A). The observed ΔV_{OC} of 200 mV correlates well with the difference in the anodic work functions of PEDOT:PSS (work function of -5.1 eV) and MoO₃ (see Figure 1b), assuming MoO₃ being a p-type semiconductor with a valence band at -5.3 eV, as reported by Yang et al.^[13] From the literature, a clear influence of the anodic work function on the *V_{OC}* and thus on the device performance is known.^[18] The low-lying valence band of MoO₃ enhances hole collection from the donor while the high-lying conduction band serves as an electron barrier.^[13,19] Thus, our observations correlate well with the published model of MoO₃ serving as a p-type semiconductor. Besides the *V_{OC}* increase, we observed almost identical short-circuit currents (*J_{SC}*) and a slightly reduced fill factor (*FF*) for the MoO₃ cells. The reduced *FF* could be explained by an increased series resistance *R_s* in the cell due to lower MoO₃ conductance.^[19] Overall, the prevailing *V_{OC}* increase in MoO₃ cells leads to an increase in the averaged *PCE* from 4.5% to 5.5% (see Table 1 for details). Also, it should be mentioned that replacing SOL-processable PEDOT:PSS with VAC-deposited MoO₃ makes the entire device stack VAC-processable and enables highly reproducible cell fabrication.

Further cell optimization was achieved by controlling the active layer composition and thickness. Figure 2 shows the critical cell parameters *V_{OC}*, *J_{SC}*, *FF*, and *PCE* as a function of the C₆₀ content at an optimum active layer thickness of 55 ± 5 nm. While *V_{OC}* and *FF* seem to be almost independent of the C₆₀ content, *J_{SC}* peaks at a C₆₀ content of 55%. The results of the best SM-BHJ devices on PEDOT:PSS (device A) and on MoO₃ (device B and C) are summarized in Table 1. For the optimized device we measured *PCE*s of up to 6.1% (device C). Slightly higher C₆₀ content leads to a 10% increase in *J_{SC}*, whereas the deviation in *FF* is minor and almost below the error limit. The morphology in VAC-processed HB194 solar cells with 50% C₆₀ content was previously studied by STEM.^[11] A clear phase separation with domain sizes of about 10 nm was discovered. For active layers with a slightly higher C₆₀ content (55%) we expect a very similar morphology. This view is also supported by AFM.

The remarkably high *J_{SC}* in all three devices (A–C; see Table 1) results from notably strong absorption in the wavelength range

Table 1. Device performance parameters.^{a)}The structures of the devices were: **A,D:** ITO|30 nm PEDOT:PSS|55 nm act. layer|6 nm BPhen|120 nm Ag; **B,C:** ITO|20 nm MoO₃|55 nm act. layer|6 nm BPhen|120 nm Ag.

Device	HCC	Fullerene C _{xx}	C _{xx} content [%]	V _{oc} [mV]	J _{sc} [mA cm ⁻²]	FF	PCE [%]	Highest PCE [%]
A	PEDOT:PSS	C ₆₀	50	770 ± 35	12.1 ± 0.7	0.45 ± 0.01	4.5 ± 0.4	4.9
B	MoO ₃	C ₆₀	50	969 ± 2	11.8 ± 0.5	0.49 ± 0.01	5.5 ± 0.3	5.8
C	MoO ₃	C ₆₀	55	962 ± 3	12.6 ± 0.4	0.47 ± 0.02	5.8 ± 0.3	6.1
D	PEDOT:PSS	C ₇₀	50	500 ± 35	6.8 ± 0.6	0.33 ± 0.03	1.4 ± 0.4	1.8

^{a)}The data refers to an average of at least ten independent device measurements, with the exception of the last column, which presents the highest individual measured efficiency for each device setup. The illumination intensity was 100 mW cm⁻² (AM 1.5 conditions).

from 350–700 nm and high external quantum efficiency (*EQE*) of up to 75% (see **Figure 3a**). Please note that the *EQE* remains almost constant at this high level for wavelengths below the main absorption. By multiplying the *EQE* spectrum of device **C** with the AM 1.5 solar spectrum and integrating the result over the whole wavelength range, we obtained an independent measure for the *J_{SC}* value. The calculated *J_{SC}* value equals 11.5 mA cm⁻² and is in good agreement with the measured *J_{SC}* values under the consideration of measurement uncertainties (e.g., limited wavelength range used in *EQE*). **Figure 3b** shows the *JV*-curve of the devices **C**.

In addition, we tested C₇₀ as electron acceptor in the active layer, which has shown a positive impact on the cell performance in some devices, mainly due to a higher and broader absorption.^[20,21] However, interchanging the C₆₀ with C₇₀ caused a dramatic decrease in the performance of our devices (device **D**, **Table 1**).

The dye HB194 is known to show the highest hole-carrier mobility among a series of MC dyes studied previously.^[10] In order to study the hole mobility in our device, we fabricated special devices that support only injection of holes, so-called “hole-only devices” (HODs), based on the optimized devices **B** and **C**.

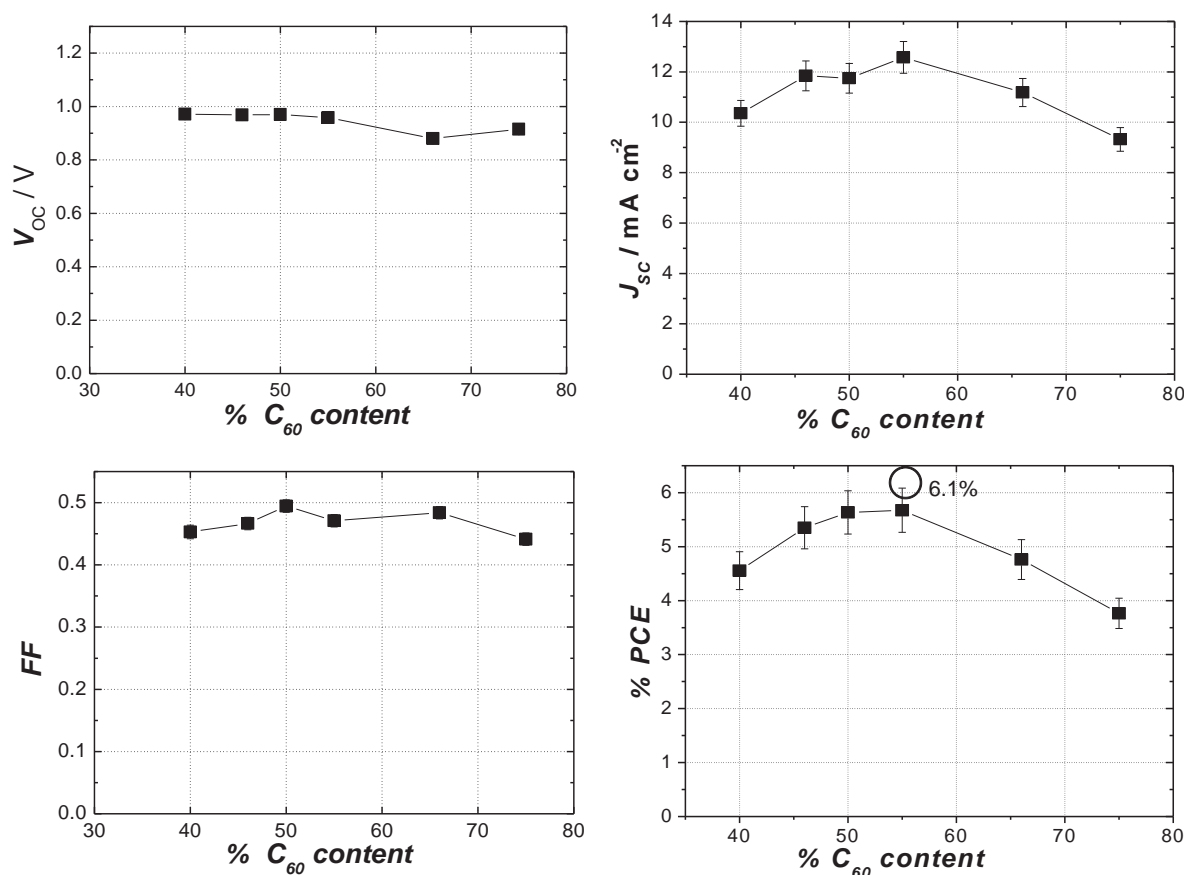


Figure 2. Averaged V_{OC}, J_{SC}, FF, and PCE of HB194 SM-BHJ solar cell (ITO|MoO₃|HB194:C₆₀|BPhen|Ag), plotted as a function of the C₆₀ content in the active layer. The optimized active layer is 55 ± 5 nm (device **C**). The efficiency of 6.1% refers to the best obtained data among at least ten independent device measurements. The line connecting the data points is a guide line for the eye. The illumination intensity was 100 mW cm⁻² (AM 1.5 conditions).

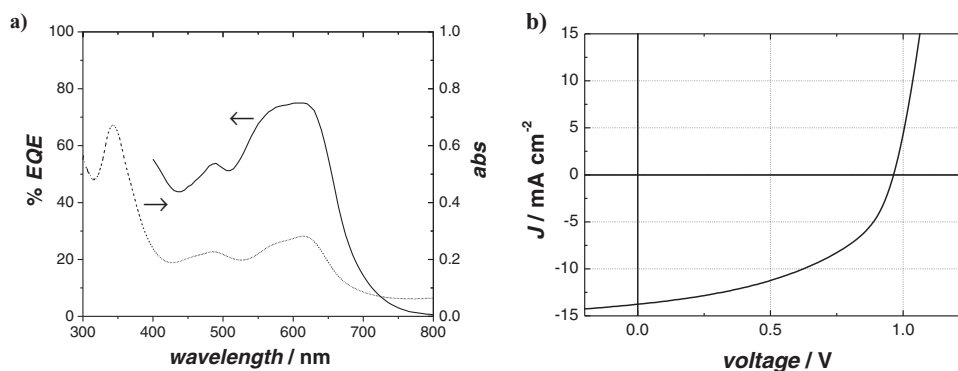


Figure 3. a) Absorption (dotted line, right axis) and EQE (solid line, left axis) spectra of device C (ITO|MoO₃|HB194:C₆₀|BPhen|Ag) with optimized layer thickness of 55 nm plotted in the wavelength range of 300–800 nm. b) *JV*-curve of the optimized HB194 SM-BHJ solar cell (device C).

By evaluation of the *JV*-data obtained from these devices using Mott's steady-state space-charge-limited current (SCLC) model, we obtained hole mobilities of $\mu_{\text{HOD}} = 2 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ in both cases. The high hole mobility is in good agreement with the observed high J_{SC} and *PCE* of the optimized devices. It is even one order of magnitude higher than the hole mobility we reported recently, which was derived from organic field-effect transistor (OFET) measurements ($\mu_{\text{OFET}} = 2 \times 10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$; 1:1 blend).^[11] Differences between these two measurements are expected, since charge transport in an OFET takes place in a mono-molecular layer atop the dielectric, which is fundamentally different from bulk transport in a HOD. However, it is quite surprising that $\mu_{\text{HOD}} > \mu_{\text{OFET}}$, as often the opposite is observed.^[22,23]

In OSCs, similar hole and electron mobilities are a prerequisite to reach high efficiency. From earlier studies by Sariciftci et al., it is known that the electron mobility in C₆₀ films is about two orders of magnitude higher than the hole mobility in HB194.^[24] Furthermore, Leo et al. have reported a reduction of the electron mobility by about half an order of magnitude in 1:1 donor/acceptor blend layers.^[25] Therefore, we can assume that the electron mobility approaches the hole mobility in our devices. Nevertheless, the limited *FF* of 0.45–0.50 (Table 1) seems to result from a comparably low hole mobility. In order to further improve the *FF* and thus the device performance, the hole mobility in the MC needs to be improved.

Finally, we studied the performance of the optimized HB194 SM-BHJ solar cell under various light intensities from 10–300 mW cm⁻² (Figure 4). It becomes obvious that even at low light intensities the cell efficiency is still high, at $(6.0 \pm 0.2)\%$. We observe a slight decrease in the V_{OC} from 960 to 910 mV with a slope of 0.06 V from standard illumination to lower light intensities. From the fit to the experimental data we determined (nkT/e) according to the following equation, when assuming a linear dependence of J_{SC} on the light intensity

$$V_{\text{oc}} = \left(\frac{nkT}{e} \right) \ln \left(\frac{J_{\text{sc}}}{J_{\text{s}}} + 1 \right) \quad (1)$$

where n is the ideality factor, k is the Boltzmann constant, T is temperature, e is the elementary charge, and J_{s} is the (reverse bias) saturation current density. This leads to an ideality factor

of $n = 0.96$, when setting T to room temperature (298 K). Hence, our devices seem to show an almost ideal diode behavior. In the literature, polymer/fullerene solar cells with a typical ideality factor n of 1.4 have been reported,^[26] indicative of recombination losses in the device (when $n > 1$).^[27,28]

The light intensity-dependent measurements further confirm the above assumption of a linear J_{SC} dependence. From our experimental data, we obtain a slope of 1.02 (see Figure 4). For higher light intensities we observe a slightly sublinear relation (slope of 0.78), possibly indicating the occurrence of a space-charge-limited (SCL) current.^[29] Based on previous work by Blom et al. the occurrence of an SCL current has a negative impact on the device performance.^[26,30]

The *FF* increases from 0.48 to 0.56 towards lower light intensities with a slope of 0.07. The higher *FF* at low light intensity could be explained by an increase in the parallel resistance (R_{p}) towards lower light intensities, as shown in previous studies by Hummelen et al.^[29] In general, *FF* depends on R_{p} as well as R_{s} .

3. Conclusions

We successfully optimized a simple layer stack SM-BHJ solar cell based on the MC dye HB194, achieving *PCEs* up to 6.1%. Introducing MoO₃ as HCC resulted in an increase of V_{OC} from 770 to 970 mV. Optimizing the composition with slightly higher C₆₀ content in the BHJ led to a further increase in J_{SC} from 11.8 to 12.6 mA cm⁻². EQE measurements show a clear peak around the wavelength of maximum light absorption ($\lambda_{\text{max}} \approx 600 \text{ nm}$). The high EQE of up to 75% supports the observation of remarkably high current densities in the SM-BHJ devices based on the dye HB194. Light intensity-dependent studies demonstrated constant (excellent) device performance throughout a large light intensity range, unlike many other systems reported in the literature.

Based on the successful introduction of MC dyes in VAC-processed SM-BHJ solar cells, we further aim at the application of MC dyes in tandem solar cells. Due to their remarkably high absorption, MC dyes are promising candidates for tandem structures. Another important advantage is that MC dyes can be easily synthesized at large scale and high purity.

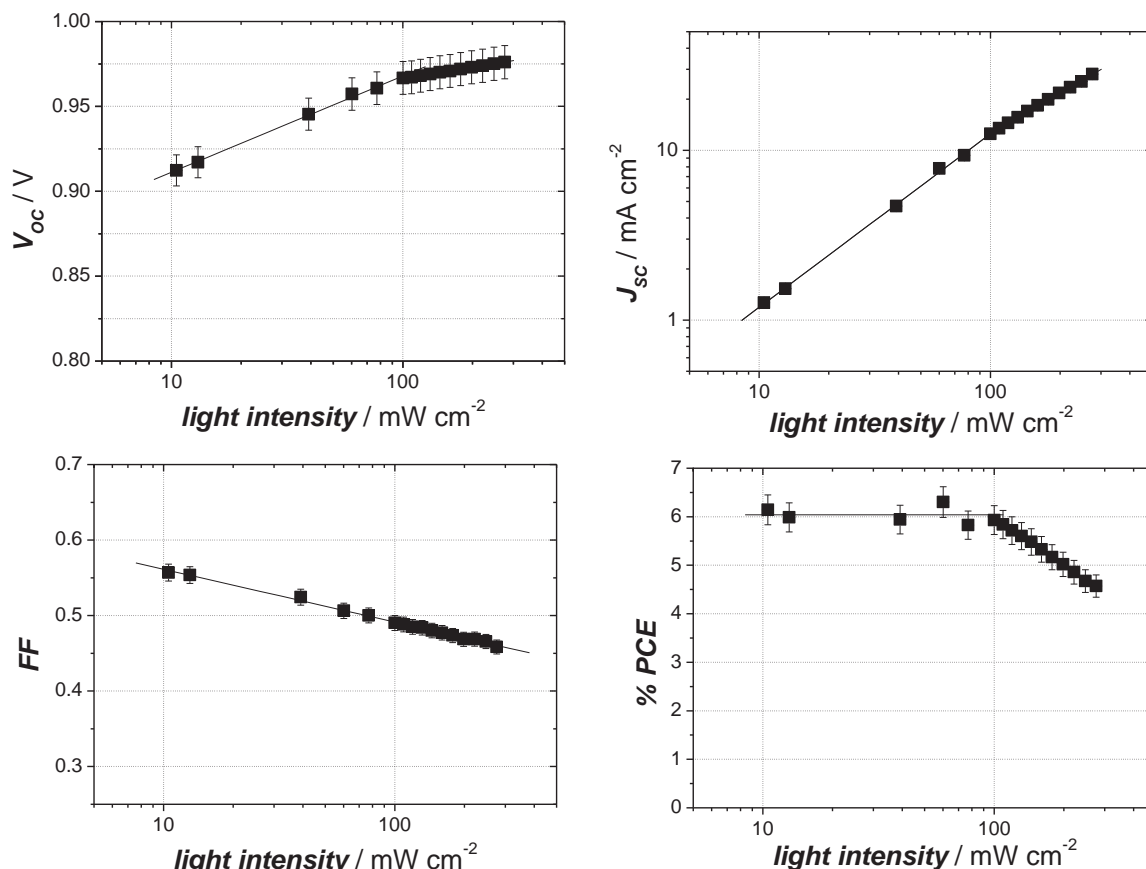


Figure 4. V_{oc} , J_{sc} , FF , and PCE of optimized HB194 SM-BHJ solar cell (device C) are shown for various light intensities from 10–300 mW cm^{-2} . The lines illustrate the slopes in V_{oc} , J_{sc} and FF as well as the averaged PCE in the light intensity range from 10–100 mW cm^{-2} .

4. Experimental Section

All devices were fabricated on commercial indium-tin oxide (ITO)-coated glass. The ITO was patterned by etching, followed by a standard cleaning process.^[11] Before device fabrication the ITO surface was exposed to ozone for 10 min. Afterwards, the ITO substrates were either coated with PEDOT:PSS (CLEVIOSTTM P AL 4083, Heraeus) and heat-treated for 2 min at 110 °C or directly transferred to the vacuum chamber for MoO_3 (99.95%, Alfa Aesar) deposition. All VAC-processed layers – except gold – were deposited in a custom-made ultrahigh vacuum chamber (K. J. Lesker Co., UK) at a base pressure of 10^{-6} – 10^{-7} mbar. The gold layer was deposited in a smaller vacuum system for reduced material consumption. By co-evaporating C_{60} (2× sublimed, CreaPhys GmbH, Dresden) and the dye HB194 the BHJ layer was fabricated. As EBL BPhen (1× sublimed, Sensient Imaging Technologies GmbH) was used. Finally, seven top silver (99.9%, Alfa Aesar) contacts with the size of 0.08 cm^2 were evaporated on top of the organic layers.

The JV-characteristics were measured at a Keithley 2425 source-measurement unit using a filtered Xe lamp with AM 1.5 filters. EQE measurements were conducted with a setup from Newport Oriel Product Line using a 300 W xenon, ozone-free arc lamp and a Newport Cornerstone monochromator. For light intensity studies, Xe lamp filters with various optical densities were applied. Layer thicknesses were determined with a surface profiler (Dektak, Veeco). The absorption spectra were measured with a Varian Cary 50 spectrometer. Based on Mott's steady-state space-charge-limited current (SCLC) model hole-only devices (ITO|20 nm MoO_3 |55 nm bulk (1:1 ratio) |6 nm BPhen|100 nm Au) were investigated.^[29] The hole mobility was determined by fitting the

experimental data with the following parameters: $\epsilon_0 = 8.85 \times 10^{-12} \text{ F m}^{-2}$, $\epsilon_r = 3.5$, $d = 55 \text{ nm}$ (experimental), and $V_{bi} = 0.9 \text{ V}$.

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