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Analysis of the degradation mechanism of ITO-free organic solar cells under UV radiation



Solar Energy Material

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ABSTRACT

This work reports on the stability of encapsulated ITO-free bulk heterojunction organic solar cells (BHJ-OSC) under UV radiation in ambient air with the layer sequence Cr/Al/Cr/photoactive layer (PAL)/ poly (3, 4-ethylenedioxythiophene) poly (styrenesulfonate) (PEDOT:PSS)/metal-grid. The PAL consisted of poly(3-hexylthiophene) (P3HT) as a donor and (6,6)-phenyl-C₆₀ butyric acid methyl ester (PCBM) as an acceptor. BHJ-OSC with this setup showed remarkable stability under continuous illumination (1000 W/ m²) with a low UV content. In contrast, the devices degraded significantly under UV radiation which was characterized by a reduction in fill factor and short-circuit current density. Additional experiments revealed an increase of the sheet resistance of the PEDOT:PSS layer which was interestingly much more pronounced in pure PEDOT:PSS samples as compared to samples where a PAL was deposited underneath. In addition, current extraction by linear increasing voltage (CELIV) measurements indicated a decrease of the effective charge carrier mobility of the PAL. Numerical simulations based on the experimentally determined parameters showed good agreement of the solar cell performance as a function of UV exposure duration. This suggests that the increase of the sheet resistance of the polymeric hole contact and to a lesser extent the change of the effective mobility of the PAL are the main factors governing the deterioration of the photovoltaic performance upon UV exposure. A comparison to devices with a setup ITO/ZnO/PAL/PEDOT:PSS and a full metallization showed clearly improved UV stability, although the absorption of UV in the PAL is very similar. This further supports our interpretation that the degradation of the PAL plays a very minor role. The issue with the degradation of the PEDOT:PSS can easily be solved by incorporating an UV-filter into the device or preferably the use of UV-stabilized PEDOT:PSS formulations.

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1. Introduction

Evidence for photo induced electron transfer from the excited state of a conducting polymer onto buckminsterfullerene [1] promoted the development of organic solar cells (OSCs). Properties like mechanical flexibility, light weight, transparency, solution processing for low cost roll to roll fabrication make OSCs attractive [2–5]. Despite of continuous improvements in efficiency [6–8] and lifetime [9,10] achieved in the last few years OSCs still lack behind compared to the other established PV technologies.

The lifetime of OSCs is a result of several processes occurring simultaneously. A more detailed understanding of the specific degradation mechanisms can help to improve the stability of OSCs. Exposure to oxygen, water, ultra violet (UV) radiation and elevated temperatures are some of the well-known causes responsible for degradation of OSCs [5]. For some polymers used as donor in BHJ-OSCs, UV radiation can result in irreversible photodegradation [5,11–12]. Madsen et al. reported reduction in the power conversion efficiency by photochemical reactions in the active layer of OSCs by shortening of the π -conjugation [13]. Rabek and coworkers found that UV radiation together with oxygen accelerates the degradation process via singlet oxygen formation [14].

This work focuses on the underlying degradation mechanisms of encapsulated indium tin oxide (ITO) free bulk heterojunction (BHJ) OSCs under UV radiation where poly (3, 4-ethylenedioxythiophene) poly (styrenesulfonate) PEDOT:PSS has been used as hole transport layer. PEDOT:PSS has high transparency, high conductivity and can form a good contact with many semiconductors but its hygroscopic nature may facilitate water vapor ingress and accelerate the degradation [15–17]. The device stack used in this study has shown excellent stability [9] under continuous

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Fig. 1. Schematic diagram of the encapsulated ITO free BHJ-OSCs used in this study (not drawn to scale)

illumination with visible light of a sulfur plasma lamp and also under fluorescent lamps. Furthermore unpublished results show that excellent stability can be achieved under high temperatures (85 °C). In contrast to these promising results the devices degraded significantly faster under full sun illumination containing UV as observed in the ISOS 3 inter laboratory study [18]. Elschner observed a reduction in the conductivity of PEDOT:PSS upon UV illumination and relates this to light induced oxidation. He also identified wavelengths below 315 nm (UV-B) to be especially harmful. Author further revealed that the process of UVdegradation can be significantly slowed down by properly protecting the films against oxygen and humidity, i.e. depositing a UV absorption layer and by employing stabilizing additives [19]. A faster degradation rate of devices with PEDOT:PSS compared to that of devices without PEDOT:PSS has been reported [20,21] and correlated with the hygroscopic nature of the PEDOT:PSS interlayer enhancing the moisture trapping process. Additionally, a loss in conductivity of PEDOT:PSS due to water uptake is also considered to be responsible for degradation of OSCs [22].

In this device setup (see Fig. 1) lateral current collection is occurring in the PEDOT:PSS layer. Thus the average transport path length for the holes in the PEDOT:PSS layer is orders of magnitude higher (1 mm) compared to ITO based device setups (e.g. ITO/ PEDOT:PSS/photoactive layer (PAL)/LiF/Al or ITO/ZnO/PAL/PEDOT: PSS/Au), where the current must only vertically pass through the PEDOT:PSS layer (< 100 nm). Thus, the conductivity of the polymeric hole transport layer must be orders of magnitude higher for the performance of the devices under study as compared to ITO-based devices. Therefore the degradation of the PEDOT:PSS conductivity was a very probable cause for the observed instability in the ISOS-3 study and in this work the effect is quantified.

2. Experimental

ITO free BHJ-OSCs with the layer sequence as shown in Fig. 1 have been fabricated [9]. Chromium/aluminum/chromium (5 nm/100 nm/ 5 nm) layer as an electron contact has been evaporated (e-beam evaporation for chromium and thermal evaporation for aluminum) on a glass substrate in high vacuum followed by spin coating of the PAL (220 nm) under inert atmosphere. The solution for the PAL deposition consisted of poly(3-hexylthiophene) (P3HT) as a donor and (6,6)-phenyl-C₆₀ butyric acid methyl ester (PCBM) as an acceptor with concentrations of 20 mg and 14 mg respectively dissolved in 1 ml of O-xylene. Spin coating of the PEDOT:PSS layer (85 nm, Heraeus Clevios F010) as the polymeric hole contact has been done under ambient conditions followed by annealing under inert conditions at 85 °C for

15 min. Finally, a gold grid (100 nm) for current collection was thermally evaporated in high vacuum. Post annealing of OSCs at 120 °C for 10 min under inert conditions resulted in a further improvement of the performance. The active area of every cell was 1.1 cm². The finished devices have been encapsulated using UV curing epoxy glue (DELO LP655) and a 1 mm thick glass plate. A thin PEDOT: PSS layer was taken in order to optimize the light harvesting. Glass was chosen as encapsulation for mainly practical reasons. It is easy to apply and provides excellent barrier properties against oxygen and water. Furthermore in rooftop and façade applications glass is also a very realistic option for real world products.

In addition to complete OSCs, specific devices to determine the sheet resistance of the PEDOT:PSS layer were prepared. The glass substrates carry gold contacts in four point geometry. Samples with and without a PAL underneath the PEDOT:PSS have been prepared by spin coating. These samples have been encapsulated in the same way as the complete OSCs.

The encapsulated OSCs and PEDOT:PSS samples both with and without encapsulation have been subjected to UV radiation at ambient conditions at approximately 45 °C and 6% relative humidity. A Solaronix SA device with Philips UV-10 TLD 18 W [23] lamps has been used (wavelength from 340 to 400 nm, peak wavelength 360 nm). The intensity was similar to the one of the UV-part of the AM1.5G spectrum.

Current–voltage curves (*IV*) and current extraction by linear increasing voltage (CELIV) [24,25] were recorded in regular intervals for OSCs to determine the photovoltaic parameters and the effective charge carrier mobility. In the same intervals the sheet resistance of PEDOT:PSS was measured with the four-point samples throughout the ageing period.

Current–voltage (*IV*) measurements have been carried out using a Keithley 2400 source meter in four point configuration under simulated AM1.5G illumination (1000 W/m²), corrected for spectral mismatch. CELIV measurement was based on a voltage ramp applied in the dark ranging from 0.3 V to 1.2 V with duration of 30 μ s. The current has been measured with an operational amplifier (0.2 V/mA) and a Textronix TDS 2014 oscilloscope. The evolution of the sheet resistance of PEDOT:PSS samples with/ without PAL underneath exposed to UV radiation has been determined by a four point measurement with the same Keithley 2400 source meter.

For further comparison, additional ITO-based devices with the layer sequence of ITO/ZnO(zinc oxide)/PAL/PEDOT:PSS and a full metallization were fabricated. ZnO films (40 nm) were prepared by spin casting of a zinc oxide precursor (containing zinc acetylacetonate dehydrate dissolved in ethanol to a concentration of 10 mg/ml) on top of ITO structured glass substrates, followed by the annealing at 120 °C for 1 min under ambient condition. Spin coating of the PAL (220 nm, same formulation as for ITO free devices) was done under inert atmosphere and then casting of the polymeric hole contact (85 nm, Heraeus Clevios F010) was conducted under ambient condition. Devices were further annealed at 120 °C for 10 min under inert atmosphere and finally the gold contact (100 nm) was evaporated in high vacuum. These devices were encapsulated in the same way as ITO-free devices and subjected to the same ageing condition under UV radiation.

3. Results and discussion

As shown in Fig. 2 the degradation of OSCs under UV radiation is characterized by a reduction in fill factor and short-circuit current density. The rate of degradation found to be slowed down by proper encapsulation. The rate of degradation of encapsulated OSCs slowed down by orders of magnitude when a sulfur plasma lamp (ca. 1000 W/m² but with almost no UV content below Download English Version:

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