

Effects of hole-transport layer homogeneity in organic solar cells – A multi-length scale study



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ABSTRACT

Irreproducibility is a serious issue in thin film organic photovoltaic (OPV) devices, as smallest local inhomogeneities can change the entire behaviour of identically built devices without showing obvious failure. Inhomogeneities can occur at various steps of device preparation and appear in all layers with different length scales and impact. The hole-transport interlayer (HTL) in OPV devices blocks unwanted electron diffusion to the anode and corrects energetic mismatch between oxide electrode and organic semiconductor. Most commonly used is commercial ink based on poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) colloidal particles. However, exactly these are suspected to cause microscopic inhomogeneities, causing known irreproducibility of device characteristics. Considering PEDOT:PSS' acidity-caused electrode corrosion, it is questionable how much impact colloids have on device homogeneity. In this report, we give proof that a colloidal HTL does not necessarily cause device inhomogeneity and decreased efficiency, by comparing OPV devices with different HTLs, namely from commercial PEDOT:PSS ink and from MoO₃, obtained from two liquid precursors, leading to quasi-continuous or colloidal layers. With a combination of X-ray diffraction, atomic force and Kelvin probe microscopy, photoelectron and ambient air photoemission spectroscopy, we discuss the layers' properties from nano- to macroscale and demonstrate their impact upon implementation into OPV devices, via spatially-resolved characterization.

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1. Introduction

Organic photovoltaic (OPV) cells have received considerable attention as a potential source of renewable energy due to their advantages of easy fabrication, light weight, low manufacturing cost and mechanical flexibility. Typically, an OPV device consists of a transparent conductive indium tin oxide (ITO) anode, a donor-acceptor bulk-heterojunction photoactive layer and a low-work-function metallic cathode [1–4]. To enhance the collection of photogenerated charges [5] and decrease leakage currents [6] a hole-transport layer (HTL) is commonly used between ITO and the active layer.

However, despite the fact that this type of devices reaches good efficiencies (presently best known 11% [7]), a major problem of organic thin film diodes is the significant batch-to-batch and device-

to-device variation. Behaviour and performance of nominally identically built devices can differ considerably and so the research results published for identical systems. It is speculated that even device sets prepared at a time by the same experimentalist show not seldomly variations in performance of up to 10%.

Fact is that the physics of thin film diodes is naturally highly sensitive to smallest variations due to their limited lateral extension. These can be e.g. subtle differences in height, local composition, interface or contact properties, and the presence of impurities or grain boundaries. Such effects have been widely shown in the past for inorganic thin film solar cells [8]. In some cases the physical processes at such inhomogeneities were considerably altered from the expected device physics. Thereby the entire device may show a different behaviour to others of nominally identical built and not necessarily imply a clear device failure.

However, while organic devices are extensively studied on the nanoscale regarding their crucial donor-acceptor domain morphology, other rather microscopic variations originating from inhomogeneities of the electrode are often neglected. The few

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publications on this topic show its relevance, as the extensive study on organic solar cell degradation published by Hoppe et al [9], and on microscale inhomogeneities in organic solar cells and modules been published by Revière et al [10]. Both show that the effects are versatile, originating from surface roughness, work function fluctuations, activity/non-activity patches, grain boundaries, agglomerate formation, delamination, chemical reactions. And these rather intermediate-scale effects might not be visible via standard nanoprobe snapshots. In particular the poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate) (PEDOT: PSS) electrode interlayer, widely used for its high conductivity, high transparency and solution processibility [11–12] is known to generate instability and shorter life times of devices especially at exposure to humidity, due to acidity and hygroscopicity of PSS [13–14]. Additionally, originating from synthesizing the insoluble PEDOT in presence of water-soluble PSS for better handling, [15] PEDOT: PSS is forming gel-like colloidal particles, which consist of a PEDOT-rich core covered by a PSS-rich shell, [16–18]. This colloidal form is suspected to be responsible for spatial inhomogeneities and irreproducibility [19].

Here, we suggest that the local microscopic inhomogeneities in solar cells caused by the PEDOT: PSS hole-conducting interlayer are independent of the colloidal film morphology. This is demonstrated by comparison of microscopic spatial characteristics in thin film OPV devices comprising HTLs of PEDOT: PSS and of sol-gel processed MoO_3 with either colloidal form comparable to PEDOT: PSS or a quasi-continuous featureless layer.

It has been demonstrated in the past that high work-function metal-oxides, such as NiO [20–21], WO_3 [22], V_2O_5 [23–25] and MoO_3 [5–6, 25–29] might be suitable alternatives for PEDOT: PSS, especially MoO_3 , with its large bandgap between 2.9 and 3.1 eV and high work function 5.5 eV [25–26, 29–30]. In the present case, MoO_3 films are conveniently deposited from solution, equal to PEDOT: PSS. For that purpose sol-gel processing was used, allowing derivation of nano- to micron sized MoO_3 particle formulations from liquid precursors [5–6, 27–28]. Thereby the morphology, electrical properties and surface physics of the HTLs are shown with X-ray diffraction, atomic force and Kelvin probe microscopy, photoelectron and ambient air photoemission spectroscopy, and their influence on OPV device physics demonstrated, by integrated device measurements and with spatially resolved photocurrent maps from nano- to macroscale. The architecture of the studied devices is shown in Fig. 1. Herein, we use a blend of poly(3-hexylthiophene-2,5-diyl) (P3HT) and [6,6]-phenyl- C_{61} -butyric acid methyl ester (PC_{60}BM), a well-studied standard photoactive layer.

2. Experimental

2.1. Materials

Poly(3-hexylthiophene) (P3HT) was supplied by Rieke Metals Inc. (M_w 50,000–70,000 g mol^{-1} , regioregularity 91–94%). [6,6]-Phenyl- C_{61} -butyric acid methyl ester (PC_{60}BM) was purchased from Nano-C Inc. (99.5% purity). The formulation of poly(3,4 ethylenedioxythiophene): poly(4-styrenesulfonate) (PEDOT: PSS) was purchased from Heraeus Deutschland GmbH & Co. KG (Clevios P Jet (OLED)). Ammonium molybdate ($(\text{NH}_4)_2\text{MoO}_4$ ($\geq 99.98\%$)) was purchased from Sigma Aldrich. Hydrochloric acid HCl ($\geq 37\%$) and bis(acetylacetonato)dioxomolybdenum(VI) ($\text{MoO}_2(\text{acac})_2$) were bought from Sigma-Aldrich. Isopropanol was purchased from VWR International LLC. All the materials were used as received. ITO substrates (20 Ω/square , Ossila) were cleaned by sonication in acetone and isopropanol and followed by O_2 -plasma etching (100 W for 30 min) briefly before use.

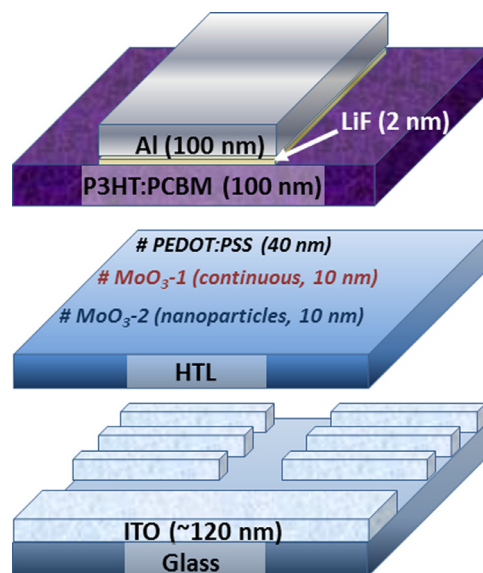


Fig. 1. Device architecture of the studied solar cells, with according layer thicknesses noted.

2.2. Preparation of the MoO_3 and PEDOT: PSS HTLs

Two different MoO_3 precursor formulations were used. The precursor leading to *continuous* films ($\text{MoO}_3\text{-1}$) was synthesized as follows: MoO_3 solution was prepared according to the procedure reported by K. Zilberberg et al. [27]. Here, $\text{MoO}_2(\text{acac})_2$ was dissolved in isopropanol to form a 0.5% (w/v) solution. The precursor formulation resulting in *nanoparticle* films ($\text{MoO}_3\text{-2}$) was prepared by hydration method in aqueous solution as reported by Liu et al. [6]. Here, $(\text{NH}_4)_2\text{MoO}_4$ was dissolved in distilled water to form a 0.005 mol/L solution. Then 2 mol/L aqueous hydrochloric acid (HCl) was added drop-wise under stirring until the pH value of the solution was between 1 and 1.5. MoO_3 precursor formulations and PEDOT: PSS, were both filtered by 0.22 μm PVDF membrane filters (Sigma Aldrich) and spin-coated onto ITO substrates at 4000 rpm for PEDOT: PSS and 3000 rpm for MoO_3 for 40 s, respectively. The layer of $\text{MoO}_3\text{-1}$ was kept at ambient air for 1 h for hydrolysis at room temperature and then annealed at 160 $^\circ\text{C}$ for 20 min. The $\text{MoO}_3\text{-2}$ film was directly annealed at 160 $^\circ\text{C}$ in air (20 min). The PEDOT: PSS anode buffer layers were annealed at 160 $^\circ\text{C}$ for 20 min under Argon (Ar) flow.

2.3. Device fabrication

Solar cells with P3HT: PCBM active layer according to the architecture in Fig. 1 were prepared using different HTLs based on continuous MoO_3 ($\text{MoO}_3\text{-1}$), nanoparticle MoO_3 ($\text{MoO}_3\text{-2}$) and PEDOT: PSS, or were prepared without HTL (= ITO). For the devices, where applicable, hole-conduction layers were deposited on patterned ITO glass substrates and treated as described in 2.2. The active layer was applied in an argon atmosphere by spin-coating from a solution of P3HT and PCBM (1:1 weight ratio, each 18 mg/mL) in 70 $^\circ\text{C}$ chlorobenzene at 2500 rpm for 60 s, followed by annealing at 120 $^\circ\text{C}$ for 10 min. The film thickness obtained is around 150 nm. The cathode was thermally evaporated as a bilayer of LiF (2 nm)/Al (100 nm).

2.4. Characterization

Material structure and film morphologies were analysed by atomic force microscopy (AFM) using two systems: For $1 \mu\text{m} \times 1 \mu\text{m}$ a Nanosurf, EasyScan2, and for $3 \mu\text{m} \times 3 \mu\text{m}$ and

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