



Inverted bulk-heterojunction solar cell with cross-linked hole-blocking layer



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ABSTRACT

We have developed a hole-blocking layer for bulk-heterojunction solar cells based on cross-linked polyethylenimine (PEI). We tested five different ether-based cross-linkers and found that all of them give comparable solar cell efficiencies. The initial idea that a cross-linked layer is more solvent resistant compared to a pristine PEI layer could not be confirmed. With and without cross-linking, the PEI layer sticks very well to the surface of the indium–tin–oxide electrode and cannot be removed by solvents used to process PEI or common organic semiconductors. The cross-linked PEI hole-blocking layer functions for multiple donor–acceptor blends. We found that using cross-linkers improves the reproducibility of the device fabrication process.

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1. Introduction

Within the field of organic solar cells, the conventional bulk-heterojunction device (see Fig. 1) comprises a substrate covered with a transparent conductive oxide, an electron blocking layer, a photoactive layer, in some case a hole blocking layer and a metal electrode [1]. Often indium–tin–oxide is used as the transparent electrode, and poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS) is applied as electron blocking layer. TiO_x, CsCO₃ or LiF are examples of hole blocking layers used in devices based on conventional design [1–4].

The photoactive bulk-heterojunction layer is located between the hole and electron blocking layers with domains of acceptor material dispersed in a donor matrix. Devices are completed with a non-transparent electrode made of a low work function metal like calcium or

aluminum. Under ambient conditions these low work function metals typically form non-conductive oxides leading to a rapid degradation of the solar cell performance [5,6]. Therefore rigorous encapsulation of these devices is required using costly packaging materials with very low oxygen and water permeation.

Inverted bulk-heterojunction solar cells (see Fig. 1) are based on an opposite layer sequence having the hole blocking layer between the transparent electrode and the photoactive layer. The top contact uses an electron blocking layer and a high work-function, air-stable material like silver or gold. The first inverted organic solar cell was prepared by Shirakawa et al. [7] applying a compact ZnO hole-blocking layer and a gold layer as top electrode. Various other oxides (such as solution processed TiO_x [8] and ZnO[9]), organic or polymeric interlayers[10] or CsCO₃[11] were reported as efficient hole-blocking layers on ITO. As electron blocking layers, PEDOT:PSS and different p-type oxides like MoO₃ or V₂O₅ have been applied [12]. Substantially improved stabilities under ambient conditions were

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reported for inverted devices while the power conversion efficiencies of conventional and inverted bulk-heterojunction solar cells made of the same absorber blend were found to be comparable. In addition, Krebs et al. demonstrated that inverted cells can be manufactured solely via solution processing making the inverted design especially interesting for large area coating [13].

The amine-rich polymer polyethylenimine (PEI) may also serve as a hole-blocking layer in organic solar cells, as published in the patent application (US2008/0264499 A1, now patent US8242356) in 2008. Recently Zhou et al. [14] reported the use of PEI as thin interlayer between the photoactive layer and an electrode material. The authors demonstrated that applying PEI on various different materials shifts the work function and leads to an electron selective contact. This system allows air-stable materials to be used in place of reactive Ca or LiF. Here we report a similar approach applying a cross-linked PEI layer on top of ITO. The motivation for this work was to show that cross-linked PEI-layers were insoluble and more robust, which may be used in solution processed multi-junction devices. Cross-linking PEI is very well known in the framework of membrane science and we have selected 5 different ether-based cross-linkers [15]. After some optimization, high performance devices were made with all of the selected cross-linkers. We found that all PEI films, with and without cross-linking, were robust to various solvent treatments. However, the developed process for cross-linked PEI interlayers is easy and insensitive to small variations in the processing conditions, which is essential for processing very thin (<10 nm) layers.

2. Experimental

Regio-regular poly(3-hexylthiophen-2,5-diyl) (P3HT) was purchased from Rieke Metals, Poly[[4,8-bis[(2-ethylhexyl)oxy]benzo[1,2-b:4,5-b']dithiophene-2,6-diyl][3-fluoro-2-[(2-ethylhexyl)carbonyl]thieno[3,4-b]thiophenediyl]] (PTB7) was purchased from 1-Material, PC60BM was purchased from SolenneBV, polyethylenimine branched (Mw = 800 g/mol) and the cross-linkers glycerol diglycidyl ether (CL1), bisphenol A diglycidyl ether (CL2), 1,4-butanediol diglycidyl ether (CL3), poly(propylene glycol) diglycidyl ether (CL4) and trimethylolethane triglycidyl ether (CL5) were purchased from Sigma Aldrich [16–19]. Materials were used as received without further purification. P3HT and PC60BM were dissolved together in chlorobenzene (1:0.7 wt%). PEI was dissolved in butanol (0.27 mg/ml). PEI and crosslinkers were also dissolved in butanol at following weight ratios per milliliter of butanol:

PEI/Glycerol diglycidyl ether (0.27 mg/0.44 mg), PEI/Bisphenol A diglycidyl ether (0.27 mg/0.74 mg), PEI/1,4-Butanediol diglycidyl ether (0.27 mg/0.44 mg), PEI/Poly(propylene glycol) diglycidyl ether (0.27 mg/0.82 mg) and PEI:Trimethylolethane Triglycidyl Ether (0.27 mg/0.42 mg). Devices were fabricated on indium tin oxide (ITO) coated glass substrates with a sheet resistance 15 Ω /square. The substrates were cleaned by sonication in acetone and isopropanol for 15 min. PEI-solution and the PEI:Crosslinker solutions were spin coated on the ITO

substrates at 2000 rpm and then annealed at 105 °C on a hotplate for 10 min. Then the blend of P3HT and PCBM was spin coated at 1500 rpm. and annealed at 110 °C for 5 min. The active layer thickness was ~80 nm. Finally, an 8 nm MoO₃ layer and a 100 nm Ag layer were evaporated through a shadow mask to define the active area (~9 mm²) and form the hole-collecting electrode. Conventional bulk heterojunction solar cells were prepared by spin-coating a 50 nm thick PEDOT:PSS layer (Clevios P 4083) on top of a cleaned glass substrates coated with ITO. The active layer was deposited and annealed as described above. The device was finished by evaporating 0.8 nm of LiF and 80 nm of Aluminium through a shadow mask. Devices were characterized on a Steuernagel solar simulator and external quantum efficiencies were measured on a home-built setup based on a white-light lamp, a monochromator and a lock-in detection. Device lifetime studies were performed by storage of the devices in ambient air conditions, with periodic measurements under white light.

Kelvin probe force microscopy (KPFM) was employed to confirm the intended changes in work function of the PEI interlayers. KPFM is a variant of atomic force microscopy (AFM) that allows mapping the local contact potential difference between an AFM tip and the sample surface [20,21]. The technique therefore yields the local work function and is also applicable to organic thin films [22,23]. The KPFM investigations were performed on an MFP-3D atomic force microscope from AsylumResearch. As probes, TiN coated NSG 03 silicon cantilevers from NT-MDT were employed. These probes feature a tip radius below 35 nm, a typical resonance frequency of 90 kHz and a spring constant of 1.74 N/m. For the TiN coated tip, a work-function of approximately 5 eV was estimated [24]. A profilometer (DektakXT, Bruker) was used to determine the layer thickness of the cross-linked PEI layers. After scratching the layer with a sharp needle the profile was scanned with the dektak tip.

3. Results and discussion

The work function of the different bottom electrodes was studied using Kelvin probe force microscopy. Typical Kelvin probe pictures of ITO and an ITO layer covered with cross-linked PEI are shown in Fig. 2. The complete set of topographies and CPD scans can be found in the Supplementary content. Average values for the contact voltage potentials of all investigated devices are summarized in Table 1. The measurements show that the applied PEI- and cross-linked PEI-layers reduce the work function of the ITO by about 400–500 mV. The consistently high V_{OC} and FF in our solar cells show that this large shift is sufficient to convert the ITO into a high performance hole-blocking electrode.

The topographies of all investigated surfaces were found to be very similar. Please note, that the CPD fluctuations in the KPFM images, presented in Fig. 2, as for the other samples investigated are in the range of 0.05 V. These fluctuations are mainly caused by slight changes of the AFM-tip properties during scanning. This confirms the high homogeneity in work function of the prepared films.

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