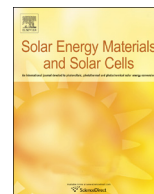




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## Improved performance and life time of inverted organic photovoltaics by using polymer interfacial materials

Zandra George<sup>a</sup>, Eszter Voroshazi<sup>b</sup>, Camilla Lindqvist<sup>a</sup>, Renee Kroon<sup>a,c</sup>, Wenliu Zhuang<sup>a</sup>, Ergang Wang<sup>a</sup>, Patrik Henriksson<sup>a</sup>, Afshin Hadipour<sup>b</sup>, Mats R. Andersson<sup>a,c,\*</sup><sup>a</sup> Department of Chemical and Biological Engineering/Polymer Technology, Chalmers University of Technology, SE-412 96 Gothenburg, Sweden<sup>b</sup> Imec-Kapeldreef, 75 B-3001 Leuven, Belgium<sup>c</sup> Ian Wark Research Institute, University of South Australia, Mawson Lakes, South Australia 5095, Australia

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## ABSTRACT

A previously published fluorene based interlayer polymer is here compared to three similar polymers where the fluorene monomer has been exchanged with monomers that have been reported to have a higher photo-chemical stability. The polymer interlayers have been studied in terms of their influence on device performance and stability on inverted devices with an active layer of P3HT:PC<sub>61</sub>BM. By acting as a hole-blocking layer the polymers are able to increase the efficiency of the devices with ~50% compared to devices with an ITO cathode. In addition, the polymers also improve the photo-stability of the devices, mainly as an effect of a reduced decrease in open-circuit voltage and fill factor. This indicates that solution processable polymer interlayers could be a way towards both higher efficiency and improved stability of inverted organic solar cells.

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## 1. Introduction

Polymer based solar cells (PSCs) are hailed for their promise of cheap renewable energy, which stems from their ability to be solution processed via cost-effective manufacturing techniques [1–4]. Additionally, these light-weight and flexible photovoltaic devices are envisioned to open up a plethora of commercial niche applications in clothing industry, portable products and off-grid markets. Until so far, the most successful polymer solar cells are bulk heterojunction (BHJ)-type devices which contain a mixture of electron donating polymer and electron withdrawing fullerene, usually [6,6]-phenyl-C61-butyric acid methyl ester (PC<sub>61</sub>BM) or [6,6]-phenyl-C71-butyric acid methyl ester (PC<sub>71</sub>BM). The performance of the devices has increased rapidly in the last years with power conversion efficiencies (PCE) now reaching ~8–10% [5–9].

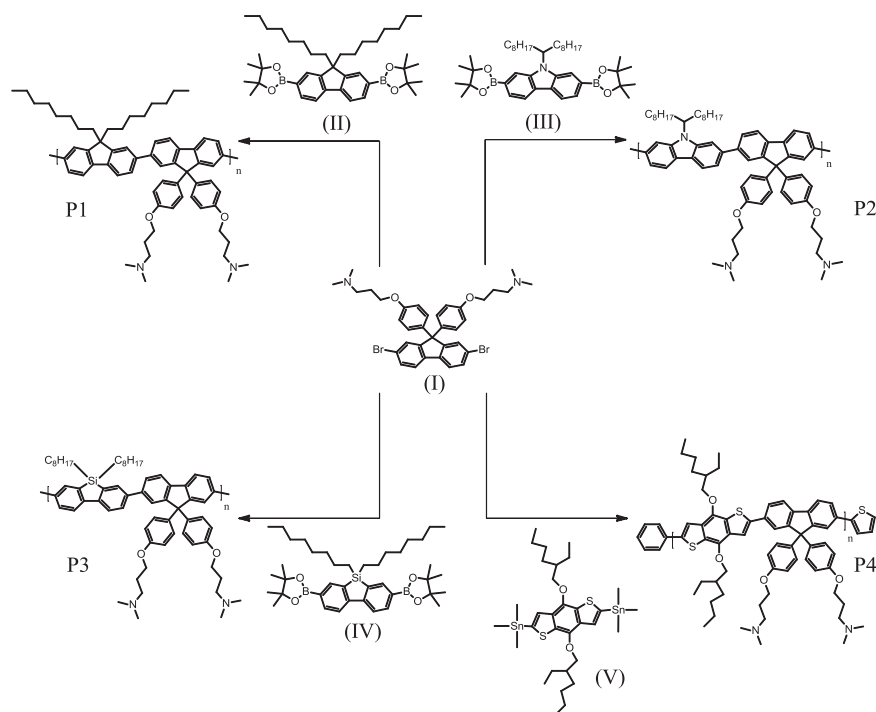
The current challenge associated with PSCs is the stability of certain devices and materials. Therefore, long-term stability has to be confirmed before commercialization is possible. To address this issue, the material layer stack employed in conventional solar cells has been inverted. These so-called inverted polymer solar cells (iPSCs) have been shown to display improved long-term stability compared to conventionally built PSCs [10,11]. On the contrary,

iPSCs often show lower PCE than conventional PSCs due to a too high work-function of the ITO cathode. To improve the electron collection efficiency (charge selectivity) at the cathode, and thereby the overall PCE, significant effort has been made to modify the interface between the cathode and the organic photo-active layer. This can be made by a coating of semiconducting metal oxides, such as zinc oxide (ZnO) and titanium oxide (TiO<sub>x</sub>) [12–16]. However, the hydrophilic surface of these materials can have a negative effect on the solar cell performance, inducing the formation of an undesired blend morphology upon deposition of the active layer [17]. In addition, organic and polymer interface materials such as polyethyleneimine (PEI) and poly(3,3'-(((9,9'-dioctyl-9H,9'H-[2,2'-bifluorene]-9,9-diyl)bis(4,1-phenylene))bis(oxy))bis(N,N-dimethylpropan-1-amine)) (PFPA-1) have been used as alternative electrode modification materials lately in order to improve the electron injection and extraction, energy level alignments and surface energy in both light emitting diodes and solar cells [18–22]. Together with their solution processability and mechanical robustness this makes the polymer interlayers a viable alternative to inorganic metal oxides [23–28].

To our knowledge, not much work has been performed on the structure–property relationship with respect to the stability of interlayer materials. Here we present three new polymer interlayer materials which are partially derived from PFPA-1 [24] (here noted as P1) (Scheme 1). The fluorene monomer has been exchanged for a carbazole (P2), a silafluorene (P3) and a benzodithiophene (P4) unit

\* Corresponding author. Tel.: +46 31 772 8227; fax: +46 31 772 3418.

E-mail address: [mats.andersson@chalmers.se](mailto:mats.andersson@chalmers.se) (M.R. Andersson).



**Scheme 1.** Chemical structures and synthesis of the polymers.

respectively since these structures have been reported to have a higher photochemical stability than the fluorene [29,30]. In addition, they will influence the energy level alignment of the materials used in devices. As reference to our previous work we also included PFFA-1, which displayed excellent performance as an interlayer molecule in iPCS using a quinoxaline-based polymer (TQ1) and PC<sub>71</sub>BM as the active layer [24,31]. The four organic interlayer polymers were evaluated both by studying device performance and stability. Additionally, comparison was made to devices with no interlayer as well as devices using TiO<sub>2</sub> as the interlayer [32].

## 2. Experimental

### 2.1. Materials

3,3'-(((2,7-dibromo-9H-fluorene-9,9-diyl)bis(4,1-phenylene))bis(oxy))bis(N,N-dimethylpropan-1-amine) (I) [24], 2,2'-(9,9-dioctyl-9H-fluorene-2,7-diyl)bis(4,4,5,5-tetramethyl-1,3,2-dioxaborolane) (II) [33], 9-(heptadecan-9-yl)-2,7-bis(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)-9H-carbazole (III) [34], 5,5-dioctyl-3,7-bis(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)-5H-dibenzo[b,d]silole (IV) [35,36] and poly(3,3'-(((9',9'-dioctyl-9H,9'H-[2,2'-bifluorene]-9,9-diyl)bis(4,1-phenylene))bis(oxy))bis(N,N-dimethylpropan-1-amine)) (P1) [24] were prepared according to literature procedures. (4,8-bis((2-ethylhexyl)oxy)benzo[1,2-b:4,5-b']dithiophene-2,6-diyl)bis(trimethylstannane) (V) was purchased from Solarmer Materials Inc. and used as received. Other chemicals, unless otherwise specified were purchased from Sigma-Aldrich and used as received. Synthetic procedures can be found in the [Supporting information](#).

### 2.2. Characterization

<sup>1</sup>H NMR and <sup>13</sup>C NMR spectra were recorded with a Varian 400/54/ASP using CDCl<sub>3</sub> as the solvent. In all cases, the peak values were calibrated relative to tetramethyl silane. Square-wave voltammetry (SWV) measurements were carried out on a CH-Instruments 650A

Electrochemical Workstation. A three-electrode setup was used with platinum wires both as working electrode and counter electrode, and Ag/Ag<sup>+</sup> used as reference electrode calibrated with Fc/Fc<sup>+</sup>. A 0.1 M solution of tetrabutylammonium hexafluorophosphate (Bu<sub>4</sub>NPF<sub>6</sub>) in anhydrous acetonitrile was used as supporting electrolyte. The polymers were deposited onto the working electrode from chloroform solution. In order to remove oxygen from the electrolyte, the system was bubbled with nitrogen prior to each experiment. The nitrogen inlet was then moved to above the liquid surface and left there during the scans. Highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) were estimated from peak potentials of the third scan by setting the oxidative peak potential of Fc/Fc<sup>+</sup> vs. the normal hydrogen electrode (NHE) to 0.630 V [37], and the NHE vs. the vacuum level to 4.5 V [38].

### 2.3. Polymer stability

The photo-chemical stability of the interlayer polymers was measured on thin polymer films spin-coated on glass. Samples were illuminated under 0.5 sun intensity with an Educational/Research benchtop open array solar simulation system from Eye Lightning International using an AM1.5G spectral matching filter and in ambient air. During illumination, the samples were placed on a black metal surface holding 50 °C (the temperature was measured with a K-type self-adhesive thermocouples from Omega, estimated error ± 5 °C). The samples were removed periodically and UV–vis absorbance spectra were recorded from 300 to 1100 nm with a Perkin Elmer Lambda 900 UV–vis–NIR absorption spectrophotometer. The maximum peak value was used to monitor the degradation of the four polymers.

### 2.4. Device fabrication

All cells have inverted structure and were fabricated onto detergent and solvent cleaned 140 nm thick, ITO coated glass substrates (Colorado concept). One device without any interlayer and one device with 3.3 nm thick TiO<sub>x</sub> buffer layer were fabricated

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