



The effect of charge extraction layers on the photo-stability of vacuum-deposited versus solution-coated organic solar cells



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ABSTRACT

Organic solar cells (OSCs) are studied for their photo-stability in inert atmosphere. Polymer solar cells with a bulk heterojunction (BHJ) of poly(3-hexylthiophene) (P3HT) and phenyl-C61-butiric acid methyl ester (PCBM) are contrasted with small molecule solar cells with a BHJ of chloroindium phthalocyanine (ClInPc) and C₆₀-fullerene. A series of charge extraction layers at the hole and electron collecting contacts are examined for their role in OSC performance and stability. The inter-compatibilities of these extraction layers in vacuum-deposited small molecule OSCs (SM-OSCs) versus solution-coated polymer OSCs (P-OSCs) are explored. Through photo-stability studies, we show that interfacial extraction layers are necessary to avoid contact photo-degradation, which otherwise leads to strong reductions in OSC efficiencies. We also highlight certain extraction layer combinations that result in strong inter-electrode degradation, and we discuss incompatibilities in extraction layers among SM-OSCs versus P-OSCs. Our results suggest that the presence of excitons at the organic-electrode interface likely plays a critical role in contact photo-degradation. By minimizing contact photo-degradation, which dominates the majority of short-term OSC degradation, a new avenue for studying OSC stability behavior and opportunities to focus on other losses in OSCs become possible.

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1. Introduction

Organic solar cells (OSCs) have achieved significant gains in efficiency in the past decade, now allowing for >10% power conversion efficiency (PCE) tandem structures [1–3]. However, in contrast, OSC stability research and the underlying degradation phenomena in OSCs are much less studied. The stability of OSCs is particularly difficult to study due to the large number of device parameters and experimental factors that may affect degradation, as well as the possibility for interplay among these factors. To this end, researchers have identified various potential avenues toward degradation, including photo-degradation [4–8], ambient (H₂O/O₂) degradation [9–13] and thermal degradation [14–16] of the photo-active organic layer. Photo-

and thermal-instability of the organic-electrode interfaces have also recently been highlighted as one of the most critical avenues toward OSC degradation [17–22]. We recently showed, by using X-ray photo-electron spectroscopy (XPS) measurements, that the organic-electrode interfaces commonly used in OSCs are inherently unstable and that they exhibit photo-induced chemical changes [20–22]. It behooves us to gain a clearer understanding of the basic role of the OSC extraction layer, and to elucidate the phenomena involved in the interfacial degradation mechanism.

The umbrella term ‘OSC’ may be broken up into small molecule OSC (SM-OSC) and polymer OSC (P-OSC). While P-OSCs are always solution-coated, SM-OSCs can be either vacuum deposited or solution-coated. Despite the different fabrication methodologies, high efficiency devices can be achieved for all of these cases [1–3], usually in combination with suitable extraction layers. It is thus intriguing that the various approaches in device fabrication

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(solution-coated versus vacuum-deposited) generally employ different extraction layers, while making use of the same electrode materials – indium tin oxide (ITO) for the anode and Al for the cathode. P-OSCs and solution-coated SM-OSCs usually employ a poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS) hole extraction layer (HEL) and a thin (~ 1 nm) LiF electron extraction layer (EEL) [23]. In contrast, vacuum deposited SM-OSCs often use a plasma treatment-based HEL [24,25] and a bathocuproine (BCP) EEL [26,27]. The relative stabilities and efficacies of these and several other extraction layers (such as CF_4 plasma treatment [25,28,29] and MoO_3 [30,31]) when directly comparing the different fabrication methodologies are, as of yet, unstudied. It follows that a comprehensive study of the inter-compatibilities and stabilities of these interfacial layers for solution-coated versus vacuum-deposited OSCs would be of substantial value.

In this work, we conduct a systematic study on the effect of commonly employed extraction layers on OSC photo-stability, as well as on the inter-compatibilities of these extraction layers in both solution-coated P-OSCs and vacuum-deposited SM-OSCs. By illuminating both SM-OSCs and P-OSCs in a controlled N_2 environment, we elucidate the impact of the interfacial layers on device lifetime and photovoltaic parameters. Our results further demonstrate that, although both SM-OSCs and P-OSCs suffer from contact photo-degradation, the use of extraction layers generally improves their photo-stability. Optimal HEL/EEL combinations are identified for both vacuum-deposited SM-OSC and P-OSC materials systems. Incompatible HEL/EEL combinations, as observed by inter-electrode degradation, are also highlighted. We suggest that the presence of excitons at the organic-electrode interface may be fundamentally important in contact photo-degradation. By minimizing contact photo-degradation, which dominates the majority of short-term OSC degradation, a new avenue for studying OSC stability behavior and opportunities to focus on other losses in OSCs become possible.

2. Experimental

In this work, both solution-coated and vacuum-deposited OSCs were fabricated on patterned ITO/glass slides. P-OSCs are comprised of a 1:1 mixture of the ubiquitous poly(3-hexylthiophene) (P3HT) donor and [6,6]-phenyl-C61-butyric acid methyl ester (PCBM) acceptor, while SM-OSCs are comprised of a 1:3 mixture of chloroindium phthalocyanine (ClInPc) donor and C_{60} /fullerene acceptor. Sublimed ClInPc was provided by the Xerox Research Centre of Canada, while other materials were purchased commercially. Detailed information regarding materials, as well as further information regarding solution preparation and substrate cleaning can be found in our previous work [20,25]. All devices were made following the standard procedure that involves the successive application of HEL, active layer, EEL and top cathode. For solution-coated OSCs, a 70 nm active P3HT:PCBM layer was formed by spincoating 1:1 P3HT:PCBM in chlorobenzene (20 mg/mL solids) at a spin speed of 1100 RPM for 60 s, followed by annealing at 110 °C for 10 min. For vacuum-deposited OSCs, the

active layer comprised a 20 nm ClInPc: C_{60} (1:3) mixed layer, followed by a 30 nm pure C_{60} layer, deposited at a total rate of 2 Å/s. All materials were deposited by thermal evaporation under high vacuum at a base pressure below 5 microtorr. For HELs: 5 nm MoO_3 films were deposited by thermal evaporation; 30 nm PEDOT:PSS films were spincoated from Clevios P VP Al4083 PEDOT:PSS at 2300 RPM, followed by annealing at 180 °C for 10 min; CF_4 plasma treatment was accomplished by exposure to $\text{CF}_4:\text{O}_2$ (3:1) plasma using a reactive ion etch system equipped with an inductively coupled plasma source at 100 W and gas pressure of 20 Pa for 2 min. For EELs: 1 nm LiF and 8 nm BCP films were deposited by thermal evaporation. A 100 nm Al top cathode was deposited by thermal evaporation to complete the devices. All tests and photo-/thermal-stability experiments were conducted in a dry N_2 environment. Photo-stability tests were carried out with 100 mW/cm² white light, with the device temperatures maintained at 40 °C. Photovoltaic parameters were measured with 1-sun AM1.5G radiation from an ABET Sun 3000 Class AAA Solar Simulator and a Keithley 2400 SourceMeter. Atomic Force Microscope (AFM) images were obtained in air using VEECO Dimension 3100 AFM in tapping mode.

3. Results and discussion

3.1. Photo-stability of organic-electrode interfaces with various interfacial layers

An illustration of the relevant devices structures for the P-OSCs and SM-OSCs are provided in Fig. 1a and b respectively. The presented device structures and comprising materials were chosen for their high degree of OSC performance reproducibility, which is essential for stability studies, and also because their initial PCE values were roughly equivalent (~ 2 – 2.2% PCE for optimized HEL/EEL), allowing for simpler comparisons across the two different fabrication methodologies. While the acceptor materials are fundamentally similar (C_{60} versus PCBM), the donor materials for the two systems are different in both their structure and their energy levels, the latter illustrated in Fig. 1c [29,32–37]. As such, the present work considers two very different OSC systems – in fabrication methodology, constituent materials and device structure – to elucidate possible commonalities regarding their stability. As will be proven in the body of this work, in spite of these differences, both of the examined SM-OSCs and P-OSCs show strong similarities in their device stability that are largely associated with photo-induced organic-electrode interfacial degradation.

Both P-OSC and SM-OSC samples were continuously illuminated over a period of 84 h in a dry N_2 environment. The photovoltaic output parameters were measured at fixed time intervals during this period. In order to gauge any degradation simply due to the storage of the devices (i.e. device shelf life), identical samples were also fabricated and kept in the dark in a dry N_2 environment and measured at the same time intervals. The initial photovoltaic parameters for representative SM-OSC and P-OSC samples (prior to illumination) are detailed in Tables 1 and 2. It

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