



Photodegradation of Si-PCPDTBT:PCBM active layer for organic solar cells applications: A surface and bulk investigation



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ABSTRACT

The photooxidation of a polymer blend film used in efficient solar cells based on poly[(4,40-bis(2-ethylhexyl)dithieno[3,2-b:20,30-d]silole)-2,6-diylalt-(2,1,3-benzothiadiazole)-4,7-diyl], (Si-PCPDTBT) and [6,6]-phenyl-C71-butyric acid methyl (PC70BM) has been investigated. A set of experiments from complementary techniques was developed to monitor the modifications during ageing that occur not only in the bulk but also at the surface. The surface analyses were performed by X-ray photoelectron spectroscopy (XPS) and atomic force microscopy (AFM), and the bulk analyses by UV-visible spectroscopy and infrared (IR) spectroscopy. The results reveal that the silicon bridge atom is the first target of degradation. We have identified the existence of a photooxidation profile within the 300 nm thick film. Such a heterogeneous degradation was confirmed by time-of-flight secondary ion mass spectrometry (TOF-SIMS) depth profiling.

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1. Introduction

Organic photovoltaic (OPV) is a promising and complementary technology to classical PV, due to its convenient integration into the urban environment [1]. New properties such as transparency, low weight and flexibility give to the OPV devices a large adaptability.

Recently, a new low band gap conjugated polymer, poly[(4,40-bis(2-ethylhexyl)dithieno[3,2-b:20,30-d]silole)-2,6-diylalt-(2,1,3-benzothiadiazole)-4,7-diyl] (Si-PCPDTBT) has been proposed for making efficient organic solar cells (OSCs) devices. [2–7] This polymer was compared to the poly[2,6-(4,4-bis(2-ethylhexyl)-4H-cyclopenta[2,1-b;3,4-b0]dithiophene)-alt-4,7-(2,1,3-benzothiadiazole)], the C-PCPDTBT version. By changing the bridging atom from C to Si, significant improvements of the device performance have been observed. The Si-polymer is a more crystalline material than the C version which is suitable for better charge transport

when blended with a fullerene derivative [6]. Additionally, the introduction of the Si atom induces a larger and more well-defined phase separation between the polymer and the fullerene which improves the charge generation and decreases the charge recombination [3]. Therefore, the performances obtained with the Si-PCPDTBT active layer are better than in the case of the carbon analogue, either with or without additive processing [8].

However, raising performances is of no use for the commercialization of the OPV technology if the stability is not maintained or increased in parallel. Depending on the OSCs application, the active layer is submitted to various sources of degradation such as light, oxygen, water and mechanical stress. While oxygen and water exposure can be reduced by packaging the devices with barrier materials, it is impossible to completely avoid oxygen diffusion during the envisaged life time of twenty years, especially when the encapsulation is supposed to be flexible and low cost. Therefore, resistant organic materials and a better understanding of the photooxidation mechanism are required. The photodegradation of various conjugated polymers or blends have been reported in the literature [9–17].

In the present work, we have investigated the photochemical

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behavior of the Si-PCPDTBT:PC₇₀BM (1:2 wt ratio) active layer under photooxidative conditions (air, AM1.5). Using various complementary techniques, such as X-ray photoemission spectroscopy (XPS) and atomic force microscopy (AFM) as surface sensitive methods as well as UV/Vis and IR spectroscopy as bulk sensitive methods, it has been possible to probe and evaluate the degradation at different stages from the irradiated surface to the bulk of the deposit. The main highlight evidenced by the combination of these analyses is the existence of a steep oxidation gradient between the surface and the bulk. ToF-SIMS which is one of the most efficient tools to perform depth profiling on a nanometric scale confirmed very clearly the existence of a photodegradation gradient. The results show the higher fragility of the active layer surface thus opening up a discussion on the origin of this degradation profile and the crucial role it plays in device degradation.

2. Experimental

2.1. Film preparation and degradation

Solution of 1% (w/v) of Si-PCPDTBT:PC₇₀BM (1:2) in orthodichlorobenzene (ODCB) was prepared and was stirred at 120 °C over night. Films of around 300 nm were deposited on ITO/glass substrates by doctor blading for AFM, XPS, UV-vis and ToF-SIMS investigations. For the IR study, the films were made on KBr substrates (transparent substrate). The films were then annealed in a glove box at 120 °C for 10 min. Light degradation was performed in ambient air (45 °C, 10%rh) with a metal halide lamp (AM 1.5, 100 mW cm⁻²).

2.2. Thickness and AFM measurements

The thickness of the samples was measured by profilometry using a KLA Tencor Alpha-step IQ apparatus. A Nanoscope IIIa atomic force microscope (AFM) from Veeco Instruments was used for surface topography measurements.

2.3. XPS measurements

XPS measurements were carried out using a multi-chamber UHV-system (base pressure 2.10⁻¹⁰ mbar), equipped with a SPECS Phoibos 150 cylindrical hemispherical analyzer and a Al K α source (1486.74 eV).

2.4. FTIR and UV-visible measurements

FTIR spectroscopy was performed using a Bruker Vertex 70 V with a base pressure of 1 mbar. Absorption spectra were recorded with a Perkin-Elmer Lambda 35 UV/vis spectrometer.

2.5. ToF-SIMS measurements

Ion ToF (SIMS IV) in dual beam mode was used to perform depth profiling. A liquid metal ion gun Bi⁺ at 25 keV was used as analyzer gun and Cs⁺ at 1 keV as sputtering gun with 300 μ m \times 300 μ m size of the scanned area. Additional information are available in a recent article.[18].

3. Results and discussion

A series of Si-PCPDTBT:PC₇₀BM thin films with similar initial absorbances (1.0 ± 0.1 at λ_{\max} , see Fig. 1) was prepared (see experimental part). The absorption spectrum of the blend consists of a broad absorbance in the UV-visible spectral region. Fig. 1 also shows the absorption domain of pure PC₇₀BM and SI-PCPDTBT polymer. We note the broad absorption domain of the PC₇₀BM and the two main bands of the polymer located at 694 and 764 nm.

Subsequently, the samples were exposed to photooxidative conditions (AM1.5 in air). As the two materials absorb directly the irradiated light and are in contact with oxygen, both the polymer and the fullerene derivative are the target of chemical changes once exposed to these irradiation conditions. In a first part of this study, we investigate the degradation related to the bulk. A second part will be dedicated to the degradation occurring at the surface of the thin film.

3.1. Bulk investigation

A direct consequence of the exposure to light and oxygen is the photo-bleaching of the polymer blend films (Fig. 2). The evolution of UV-visible absorbance decay proceeds in two steps: a first rapid decrease (0–20% of UV-visible absorbance loss) followed by a slower linear decay. This is in good agreement with a recent study devoted to the photodegradation in air of the C-PCPDTBT and the Si-PCPDTBT polymers [17]. The loss of absorbance in two steps was observed only for the Si-PCPDTBT polymer suggesting a specific mechanism of degradation for this polymer.

IR spectroscopy in transmission mode is one of the most powerful techniques to probe the evolution of the chemical

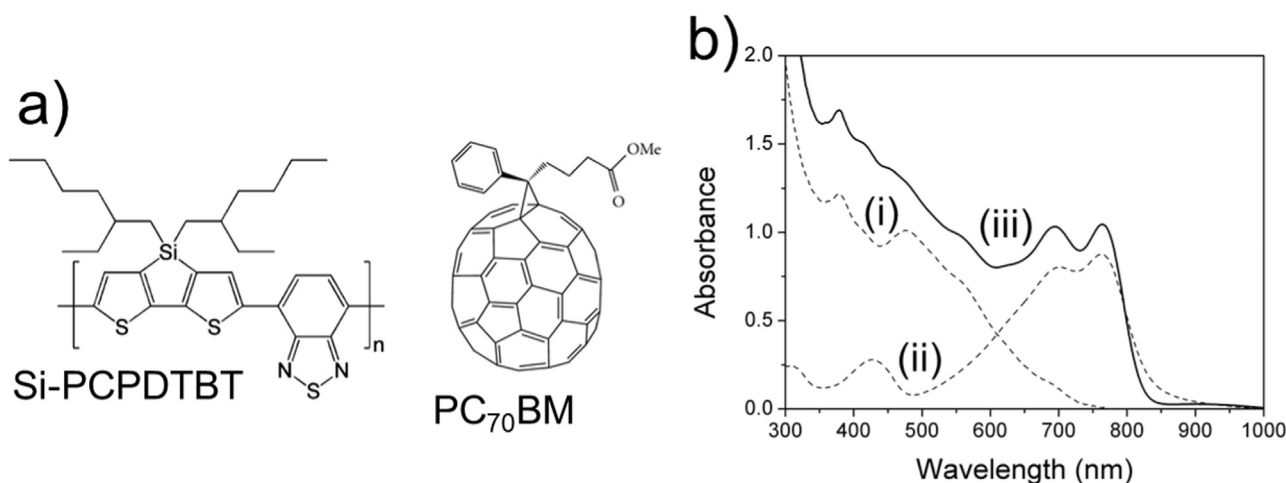


Fig. 1. a) Chemical structures of Si-PCPDTBT and PC₇₀BM and b) absorption spectra of pure PC₇₀BM (i), Si-PCPDTBT (ii) and Si-PCPDTBT:PC₇₀BM (1:2) (iii).

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